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Economic Geology

and the

Bulletin of the Society of Economic Geologists

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ECONOMIC GEOLOGY

VOL. XXXIII DECEMBER, 1938 No. 8

NOTES ON THE CHEMISTRY OF ORE SOLUTIONS.[1]

O. C. SCHMEDEMAN.

ABSTRACT.

The assumptions and evidences upon which the present "alkaline" theory of ore solutions is based are briefly summarized. A number of detailed criticisms of this theory are presented from the viewpoint of wall rock alteration. The tentative conclusion is reached that there is impressive evidence from a number of mineral deposits that ore solutions are originally acid and become neutral or alkaline only after long travel and recognizable reaction with their channelways. It is proposed that "acid" minerals develop in near-source environments.

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INTRODUCTION.

THE theory of ore deposition now widely accepted, represented largely in the writings of Lindgren, is based on the fundamental

[1] Condensed from a portion of a dissertation on "Interrelated Problems of Volcanic Activity and Ore Genesis," Harvard University 1937.

assumption that the responsible solutions are normally of alkaline reaction, and similar to those that are encountered in present-day hot springs. To the writer, this doctrine represents a pyramiding of partial evidences which, as a unit, fail adequately to account for certain features of rock alteration and the presence of "acid" minerals in ore deposits.

It is the aim of this paper to survey the nature of the assumptions and evidences upon which that theory rests, and to suggest alternative interpretations, especially in regard to certain types of wall rock alteration. The chemical character of ore solutions is fundamental and vital both to the practitioner and the theorist in mining geology, and it is to be hoped that the challenge of the problem will in the near future provide quantitative information on rock alteration in important mineral districts as a basis for sound conclusions on the nature and changes of the responsible solutions.

ACKNOWLEDGMENTS.

In preparing these notes I have drawn freely from numerous sources in the literature of ore deposits, piecing together facts and opinions published by many observers, to all of whom, but particularly to Waldemar Lindgren my debt is great. The field investigations upon which portions of this study are based were made possible in a large measure through the generosity and hospitality of the officials of various mining companies, to all of whom I extend my most sincere thanks and appreciation. The critical suggestions of fellow geologists Samuel Bowditch and Paul Henshaw during the summer trip of 1936 are also remembered with pleasure and gratitude.

The somewhat unorthodox views expressed have developed out of stimulating discussions with Professor L. C. Graton, who, though not concurring in many of my conclusions, has greatly aided the investigation through his sympathetic understanding, his pertinent criticisms, and his steady encouragement. To him, I am especially indebted and sincerely grateful.

Financial aids from Harvard University made possible my seeing the Braden mine during the summer of 1935 and many

mining districts in the Western States, Mexico and Canada in 1936.

DEFINITIONS.

Chemists define the acidity and basicity of solutions in terms of "pH" or hydrogen-ion concentration. Geologists, however, are not in a position to measure this property in ore solutions, nor are they able to predict the net result of the ionizations at high temperatures of the many possible combinations of salts in such solutions; in fact, even the chemists know little of the properties of simple salt solutions at temperatures over 100° C. To avoid controversy on this subject, an acid ore solution is considered in this discussion as a solution containing one or more of the following strong acids: hydrochloric, hydrofluoric, or sulphuric. An alkaline solution is defined as one containing no free strong acid, though it may contain one or more of the weak acids: silicic, hydrosulphuric, carbonic, or boric. This rather loose distinction appears to be the intent of the majority of those who have used the terms "acid" and "alkaline" in connection with ore solutions, for it is evident that the so-called "alkaline" hot springs and ore solutions that contain *abundant* free carbonic and hydrosulphuric acids are not strictly alkaline.

HISTORY AND ASSUMPTIONS OF THE ALKALINE VIEW.

The older concept of the importance of "mineralizers" in the formation of ore deposits, which is still held to some extent in Europe,[2] has been ascribed little importance in America during the past fifty years. Emmons [3] and Becker [4] proposed the theory that the normally insoluble sulphides are transported in ore solutions as soluble double alkaline sulphides, while Lindgren [5] called

[2] *Cf.* Vogt, J. H. L.: Magmas and igneous ore deposits. ECON. GEOL., 21: 207–233, *et seq.*, 1926.

[3] Emmons, S. F.: Geology and ore deposits of Leadville. U. S. G. S. Mon. 12, 1886.

[4] Becker, G. F.: Natural solutions of cinnabar, gold and associated sulphides. Am. Jour. Sci., 33: 199, 1887.

[5] Lindgren, W.: Metasomatic processes in fissure veins. Tr. A. I. M. E., 30: 1900. (Conclusions, page 690.)

attention to the fact that the development of sericite (which usually involves the addition of some potash) was the characteristic alteration in ore deposits. It was found that the sulphides of Hg, Au, Fe, As and Sb can be dissolved in combination with Na_2S or K_2S, which, of course, necessitates an alkaline, or very weakly acid solution. The other metals, although they form fine colloidal double sulphides, are not truly soluble under simple laboratory conditions in such combinations.[6] The difficulty of transporting the metals as sulphides in any other known manner led in large measure to the development of the theory of colloidal transport, which is best accomplished in alkaline solutions.

Geologists early found that hot spring waters are generally free from strong acids, and are hence in a proper state to transport the metals as double alkaline sulphides as advocated by Emmons, and as sulphides as proposed by the colloid theory; in fact, Becker observed cases of natural alkaline waters depositing sulphides. It was also noted that the principal components of these natural waters are alkaline carbonates, chlorides and sulphates. The presence of the chloride and sulphate radicals is of much ultimate genetic significance.

Following the lead of Lindgren, most geologists now consider that the hot spring waters are essentially the same as those which form the epithermal ore deposits, while many [7] have extended this linkage and have connected the epithermal ores with those of the deeper zones. In a strictly genetic way this is probably true, but it is highly probable that the fundamental chemistry of these solutions changes greatly as they are traced backwards towards the magma source.

Bowen [8] and Fenner [9] have recently stressed the importance and theoretical necessity of the ore solutions being initially of the

[6] Freeman, H.: The genesis of sulphide ores. Eng. and Min. Jour. Press, 120: 973–975, 1925.

[7] *Cf.* Graton, L. C.: The hydrothermal depth zones. Ore Deposits of the Western States, Lindgren Memorial Volume. A. I. M. E., 1933, pp. 181–197.

[8] Bowen, N. L.: The broader story of magmatic differentiation briefly told. In Ore Deposits of the Western States, A. I. M. E., 1933, pp. 106–128.

[9] Fenner, C. N.: Pneumatolytic processes in the formation of minerals and ores. Idem, pp. 58–106.

same character as the emanations from volcanoes. Lindgren,[10] expressing the view of some geologists, has recognized the position taken by Bowen and Fenner, and though he agrees that the solutions may be acid when they leave the magma, they: "rapidly become neutral or alkaline (through reactions with their walls) and remain thus to the surface." He also states: "The writer cannot agree with Dr. Bowen when he says (p. 127) that 'most ore deposits are formed in depth through the agency of acid solutions.'" In other words, the view of such observers is that, though the solutions may at one stage be acid in reaction they are rapidly neutralized and are not acid when they deposit ore, *i.e.*, they are not acid within our range of observation. Certainly it would seem, however, that if the original magmatic derivatives are acid, there should be abundant evidences of the fact in wall rock alterations, particularly where great quantities of solutions have been active, and in deposits formed close to the magma source.

THE INTERPRETATION OF WALL ROCK ALTERATION.

Introduction.

It has long been recognized that sericite and quartz are the two principal products of rock alteration in association with ore deposits, and that these commonly grade outwards (in the same deposit), or upwards (lower intensity zones) to a chlorite and carbonate type of alteration. From this observation it has been concluded that: potash and silica (with lesser lime and magnesia) are the main non-metallic additions from the magma; that the solutions must be both liquid and alkaline in order to carry them; and, third, the low vapor pressures of these constituents[11] makes it unlikely that they could be given off by the magma as gases within the commonly accepted range of temperatures applicable to the process. It generally is admitted that silica may escape from the magma as easily volatile chlorides or fluorides, but it is

[10] Lindgren, W.: Succession of minerals and temperatures of formation in ore deposits of magmatic affiliations. A. I. M. E. Tech. Pub. 713: 6–7, 1933.

[11] *Cf.* Tables compiled by J. H. L. Vogt: *Op. cit.*, p. 218.

commonly believed that these halogens are of insufficient quantity to account for the total amount of introduced silica often observed in ore deposits.[12]

The relative scarcity of minerals with radicals of the strong acids has also been noted, and taken as confirmation of the generally alkaline aspect of the solutions and of the alteration. The anomalous, and not uncommonly abundant, presence of the fluorine (and occasional chlorine) minerals is generally disregarded, but the sulphate minerals are recognized as out of place in the alkaline scheme of things. Their presence is considered a consequence of one or another local condition which produces sulphuric acid out of the components of alkaline solutions.[13]

The Potash Problem.

Must the potash commonly added to wall rocks by hydrothermal solutions be entirely or even partially of direct magmatic origin, as is usually assumed?[14] A suggestion that this is not necessarily the case is furnished by the *alteration zoning* so well developed in the Climax molybdenum deposit at Climax, Colorado.[15] The limits and form of the zones are partially shown in Fig. 1, and the composition of each of the zones is given in Table I.

A typical cross section of the ore deposit (Fig. 1) shows that the zones of potash addition grade *inward* and *downward* to the core of alkali-leaching. The loss of potash per unit volume in the core rock is 6.15 weight units, while the gain in the moderately

[12] *Cf.* Lindgren, W.: *Op. cit.*, Tech. Pub. 713, p. 7.

[13] Cf. Page 807 *et seq.*

[14] *Cf.* Lindgren, W.: Mineral Deposits, 4th Ed. 1933, p. 124. Lindgren, W. and Ransome, F. L.: Geology and ore deposits of the Cripple Creek district. U. S. Geol. Surv. Prof. Paper 54: 225, 1906. Lindgren, W.: *Op. cit.*, Tech. Paper 713. In this recent exposition of his views, he does not mention the potash problem, but stresses the difficulty of the derivation of silica as a gas from the magma. This omission gives the reader the impression that potash (whose compounds have a much lower vapor pressure than those of silica) no longer offers a problem. Whether or not we may interpret this as meaning that Lindgren believes that the potash may be derived from the wall rocks through leaching is another question.

[15] Butler, B. S. and Vanderwilt, J. W.: The Climax molybdenum deposit. U. S. Geol. Surv. Bull. 846–C, 1933.

silicified zone is 1.47 units, and that in the slightly silicified zone is 1.90 units. If observations were confined to the surface alone (in this diagram), the core would not be apparent, and it could only be concluded that potash had been added to the granite throughout in large quantities, probably from the underlying

TABLE I.

QUARTZ-SERICITE-ORTHOCLASE ALTERATIONS.

	Climax*				O. K.	
	1	2	3	4	5	6
SiO_2	70.83	78.62	80.78	98.23	64.41	66.87
Al_2O_3	14.41	10.80	9.26	—	15.85	18.14
Fe_2O_3	.35	.05	tr.	—	1.92	1.36
FeO	2.94	1.13	1.34	—	2.52	1.06
MgO	.56	.05	.10	—	1.66	.68
CaO	.64	.05	.10	—	3.71	.11
Na_2O	2.44	.76	.31	—	3.60	.61
K_2O	6.21	8.11	7.68	.06	3.46	4.12
H_2O-	.04	.10	.01	—	.12	.87
H_2O+	1.34	.43	.46	.02	1.09	4.05
TiO_2	.24	.04	tr.	—	.43	.85
P_2O_5	.15	.10	.08	—	.23	.05
Cl	.05	.04	.04	tr.		
F	.00	.00	.05	tr.		
S	.01	.06	.03	.01	None	.23
BaO	.02	.03	.01	—	—	—
Mo	.00	.10	.12	.01		
CO_2					.72	None
SO_3					None	.05
MnO					.07	None
Cu					—	.70
	100.22	100.47	100.37	98.52	99.79	99.75

1. Climax-granite-fresh—J. G. Fairchild, Analyst.
2. Climax-granite, slightly silicified—J. G. Fairchild, Analyst.
3. Climax-granite, moderately silicified—J. G. Fairchild, Analyst.
4. Climax-granite, highly silicified (core)—computed from thin section study.
5. O. K. monzonite—fresh—R. C. Wells, Analyst.
6. O. K. monzonite—altered—R. C. Wells, Analyst.

*Climax analyses from Bull. 846-C, p. 225. O. K. analyses from U. S. Geol. Surv. Prof. Paper 80, p. 76.

hypothetical magma source. Below the surface, however, at the level of the bottom of the diagram, there is exposed (by drilling) 3,200 feet of the "addition zones," and *1,800 feet of leached core rock.*

Calculation of the volume of the core (a cone), and the outer rings (complete conical shells), multiplied by the unit gains and losses in each form shows that, although the gain of potash in the outer and upper zones still outweighs the losses (by a ratio of 20 to 1), this is a much different condition than obtains at the surface. Still deeper, it will be seen that these full geometrical forms if projected to a depth of 2,000 feet below the bottom of the diagram would result in a core of leaching from which the

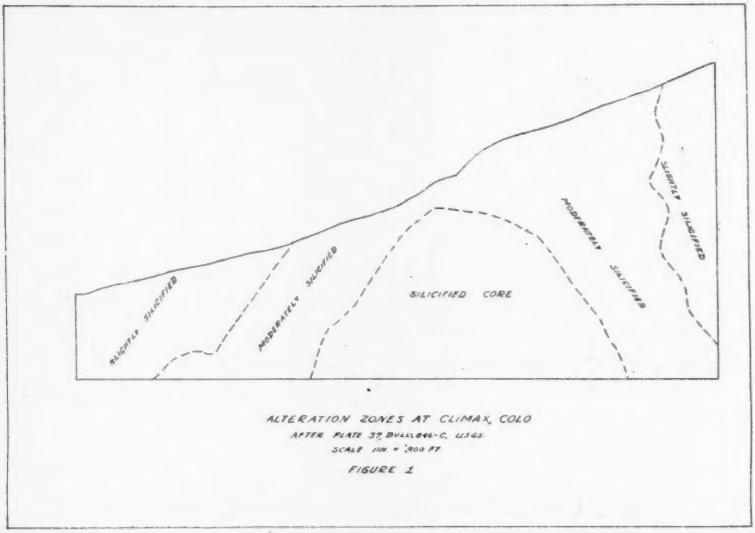

FIG. 1.

potash abstracted from that level upward would just balance that gained in the full conical shells of addition. This is speculation, of course, but it shows what the possibilities may be in the "lateral secretion" of rock constituents in the mineralization process.

The much smaller, but otherwise essentially similar O.K. pipe of Utah [16] shows the same type of zoning, and a much closer

[16] Butler, B. S.: Geology and ore deposits of the San Francisco and adjacent regions, Utah. U. S. Geol. Surv. Prof. Paper 80: 125–127, 1913.

balance between gains and losses in the portions that have been explored (Table I). It is believed that this is also the case in many replacement veins. Take, for example, a hypothetical three-foot replacement vein of sulphides encased by one-half foot sheaths of "silicified rock," *i.e.*, quartz, which in turn grade into sericitized quartz monzonite. For convenience in calculation, consider one half of this symmetrical vein. Daly[17] gives for his average quartz monzonite a potash content of 3.72 per cent (say 3.7 per cent). The loss of potash from the vein and the silicified zone would be $(1.5 + 0.5) \times 3.7$ or 7.4 units. If the sericitized monzonite contains 5.0 per cent potash,[18] then the gain per unit of rock would be 1.3, from which it can be calculated that 5.7 feet would show a gain equal to that lost in the two feet of "vein" replacement. This appears to be of about the right magnitude for the sericite zones noted next to *single* veins, though it is not true in districts such as Butte where sericitization is widespread.

An actual case of this type is given by Boutwell[19] from the Last Chance mine at Bingham. He states that the vein here varies in width from "very thin to 14 feet," but does not give a figure for the width obtaining at the point represented by his analyses. Assume, however, that the width there is 5.0 feet. The vein is of galena, sphalerite, pyrite and quartz, probably in part of open-space filling, although the descriptions are not entirely clear on this feature. The analyses of the fresh and altered rock are shown in Table II. Assuming that the intense zone of alteration obtaining directly alongside of the vein extends the two feet to the less intense zone represented by the analysis in column 2, the gain in potash in this zone is equal to

[17] Daly, R. A.: Igneous rocks and the depths of the earth, 1933, p. 15.

[18] Lindgren, W.: *Op. cit.*, p. 542, gives 5.0 per cent as the average per cent of potash in the alterations of monzonite at Clifton, Butte, Miami, Ray, Ely, Breckenridge, Colorado, and Frisco, Utah. Leith, C. K. and Mead, W. J.: Metamorphic Geology, Holt, 1915, Plate V, shows the average gain in such alterations to be of the per cent indicated by these figures, *i.e.*, 5.0/3.7—100 equals 35 per cent, as shown in their diagram. See also the data in Tables I, II and III.

[19] Boutwell, J. M.: Economic geology of the Bingham mining District, Utah. U. S. Geol. Surv. Prof. Paper 38: 178 and 276, 1905.

$2 \times (7.02 - 4.23)$ or 5.58 units. The loss from one-half the full width of the vein is equal to 2.5×4.23 or 10.6 units, providing the vein has been formed entirely through replacement. This is enough to furnish the potash gained in the intense zone plus that in $(4.91\text{-}4.23)/(10.6\text{-}5.58)$ or 13.5 feet of the less intense zone. The actual width of this less intense zone of alteration is not given by Boutwell, but to judge from his state-

TABLE II.

SERICITIZATION—BINGHAM, UTAH.

	1	2	3
SiO_2	58.64	56.17	56.78
Al_2O_3	15.35	15.94	16.90
Fe_2O_3	3.25	3.43	6.87
FeO	2.54	1.92	2.34
MgO	3.84	1.60	.03
CaO	5.37	5.19	1.18
Na_2O	3.60	2.48	.37
K_2O	4.23	4.91	7.02
H_2O-	.86	1.30	1.32
H_2O+	1.50	2.95	2.23
TiO_2	.83	.90	.81
CO_2	None	2.01	.26
P_2O_5	.02	.20	.04
S	.05	1.03	5.93
Cr_2O_3	tr.	—	tr.
MnO	tr.	tr.	tr.
BaO	.18	.18	.14
	100.26	100.21	102.22
O equivalent to S	.02	.39	2.22
Total	100.24	99.82	100.00

E. T. Allen, Analyst.

1. Fresh monzonite—British tunnel.
2. Monzonite 1–2 feet from vein—British tunnel.
3. Monzonite wall of vein—British tunnel.

From U. S. Geol. Surv. Prof. Paper 38, p. 178.

ments that the monzonite country rock in general appears quite fresh, and that only where "adjacent" to veins does it become bleached, it seems that this "less intense" zone is probably not greater than 10–15 feet in width, if it is that much. After considering the apparent gain in potash represented in this and other cases at Bingham, Boutwell was led to conclude that: ". . . the

alteration was produced by heated solutions rich in K_2O"; the assumption being that the potash was strictly hypogene in origin.

The massive sulphide body at Cerro de Pasco, Peru,[20] suggests a further example of this "lateral secretion" of potash. This body is 400 feet wide by 5,000 feet long and has formed by replacement of all of the kinds of rock that surround it. Let it be assumed that the amount of vent rocks that have been replaced is proportional to an average width of 100 feet. For rough purposes, the percentage of potash in the vent rocks may be taken as that of an average monzonite, *i.e.*, 3.7 per cent. (The amount of potash in the shaley portions of the Excelsior series would likewise be of this order, as indicated by Clarke[21]; 3.24 per cent.) One hundred feet of complete replacement in the vent rocks alone would release 370 units of potash, or enough to supply that gained in a parallel band of the vent rocks 280 feet in width; on the assumption that the average percentage of potash in these altered rocks is 5 per cent. This amount of potash is sufficient to form 42.4 per cent of sericite, which appears to be of the right order of magnitude for the average sericitized rock at Cerro. The total quantity of such sericitized rock, however, is at least double that calculated.

In all of the foregoing calculations, it is evident that the zones of potash leaching do not balance quantitatively those of addition, unless every assumption is made in favor of the lateral secretion process. However, in these cases, except that of Climax in part, the principal assumption is that the direction of travel of the potash is solely lateral. What though of the possibility that the remainder of the necessary potash is supplied from leaching in *depth?* This cannot be proved in any one district, though it is suggested at Climax. If the principle of this idea is true, the ore deposits formed in near-source environments should show more leaching than those formed farther from the source. The main difficulty here is in deciding which ore bodies are truly near-

[20] McLaughlin, D. H. and Bowditch, S. I.: The Cerro de Pasco District. Copper Resources of the World, XVI Int. Geol. Cong., *2*: 516–527, 1935.

[21] Clarke, F. W.: The data of geochemistry. U. S. Geol. Surv. Bull. 770: 34, 1924.

source in position. The deep-seated deposits may be also near-source, though this is by no means a necessary conclusion. Perhaps the best near-source examples are the pyrometasomatic deposits, and these are discussed in the following pages. A further, though controversial, type is represented by the deposits formed in the roots of volcanoes.

Lime and Magnesia.

The general role of these bases in hydrothermal alteration does not draw as much attention as that of potash, although study of the losses exhibited by these two alkaline earths in the normal type of alteration (sericite) shows that as individuals they are carried away in greater quantities than potash is gained. In other words, the general trend of the sericite type of alteration is a loss and not a gain of bases, as is implied by the emphasis placed on the addition of potash.[22]

In many ore deposits accompanied by the propylitic type of alteration either or both lime and magnesia are gained rather than lost, or at least they are lost in lesser amounts than from the sericite type of metamorphism. When the two types are found together in the same district, the propylite is always found outside of, or above the zones of sericitization. Following through the principle of "lateral secretion" suggested in the case of potash, it seems very likely that the lime and magnesia gained in the propylite zone balance those lost in the zones of silicification and sericitization, though data are not at hand to make a quantitative check of this hypothesis. The propylite type is more common in the epithermal than in the mesothermal deposits, and this appears to be an effect of major, vertical, or depth-zone zoning, the same in kind as that observed within individual districts.

The writer recognizes that magnesia and lime show little, if any, loss in the usual deep-seated gold-quartz type of rock alteration, which they should if this general theory of alteration zoning

[22] *Cf.* Lindgren, W.: *Op. cit.*, Tr. A. I. M. E., 30: 1900, p. 690. It is difficult to find a recent direct reference to this question, but the implication is everywhere evident that potash is the key constituent whose movements indicate the nature of the process and of the solutions.

is extended downward. Soda and potash likewise show little consistent change, whether gain or loss. As implied above, it is possible that these deposits are not near-source, even though they are admittedly deep-seated. It may also be that the solutions that form these deposits are from the outset of a fundamentally different nature from those that form the deposits of the shallower zones; a consequence of deeper-seated, more extended differentiation in the parent magma.

The contact metamorphic or pyrometasomatic deposits show a consistent and quantitatively great loss of lime and magnesia, and they are generally conceded to have been formed close to the magma source, though not as close as was formerly supposed, *i.e.*, while the neighboring intrusive was still molten. The second noteworthy feature of these deposits is that they exhibit large gains in silica and iron.[23] Both silica and iron form easily volatile compounds with the halogens, and silica forms a hydride compound of high vapor pressure. Their introduction in acid solutions thus offers no major *a priori* difficulty on the assumption that they separated from the magma as acid gases. The proponents of the alkaline view, however, believe that there is insufficient evidence of the activity of acid solutions in the rocks to warrant this assumption. They feel that fluorine and chlorine minerals are of too limited development in these deposits, and that there is a notable lack of excess of solution over deposition, which should be expected if free acids had been active in the alteration of the limestone host rocks. In reply to the first argument, it is true that the fluorine minerals are present in only small amounts. It is also true, though, that a small proportion of these deposits contain notable amounts of the chlorine-bearing mineral scapolite, which is found but sparingly in any other environment. The chlorine minerals are few in number and rare, so perhaps the presence of scapolite in these contact metamorphic deposits is more significant of the activity of chlorine than might be suspected. In the general case where chlorine minerals are absent,

[23] In the complete replacement of limestone to pyrite, the alteration is such as to produce a gain in bases, *i.e.*, 46.6 parts of iron are added and 40 parts of calcium abstracted.

the reason may lie in the very high solubility of the simple salts of this element. Next to the carbonates the chlorides are the most common salts in the waters of hot springs. In most cases the chlorine of these salts must surely be strictly hypogene in origin, a fact which if true, is proof of an abundance of chlorine in ore solutions. It is an open question, however, if this chlorine is ever combined as hydrochloric acid. A further indirect evidence of the activity of chlorine in ore deposition, particularly in the contact metamorphic deposits, is the frequent development in depth of large quantities of magnetite and specularite. It is possible that these minerals may form from carbonate or sulphate salts, although the hydrolysis of the chloride seems a more likely and more direct reaction.

As to the second objection of the "alkaline" view, that of the conspicuous absence of excess of solution over deposition in these deposits, little can be said; in fact, it is the writer's contention that acid attack commonly results in vugs, caves, and collapse breccias. It is noteworthy, however, that the extensive and thorough alteration of the limestones at Morococha, Peru, to anhydrite (presumably by solutions carrying free sulphuric acid) is one of strict volume-for-volume replacement as far as can be observed. In other words, the presence of free acids in hydrothermal solutions does not necessarily result in an excess of solution over deposition.

A further type of deposits believed by the writer to be of near-source position is represented by the tin and tourmaline veins and pipes in igneous rocks. These are conceded by most geologists to have formed at high temperatures though many might not agree that they were also formed close to the magma source. Analyses from four tourmaline and tin-tourmaline deposits are listed in Table IV. From these it is evident that lime and soda are lost in large proportions, while potash is somewhat decreased in amount, and magnesia, surprisingly enough, shows a tendency to remain constant. This is a result of its incorporation in tourmaline, or in biotite as at Braden, although it is quite contrary to the course followed by its usual associates—lime and soda.

TABLE III.

SERICITIZATION AND PROPYLITIZATION.

Butte, Montana.

	1	2	3	4
SiO_2	64.03	62.09	66.90	54.30
TiO_2	.60	.51	.68	.61
Al_2O_3	15.58	15.49	15.83	13.63
Fe_2O_3	1.96	{ 8.52	{ 4.37	1.89
FeO	2.83	}	Fe }	2.22
CaO	4.20	.20	.05	7.36
MgO	2.15	.42	tr.	2.13
K_2O	4.11	4.34	.03	4.41
Na_2O	2.76	.37	.08	.16
H_2O below 110° C.	.48	1.20	.87	2.19
H_2O above 110° C.	.73	3.01	3.88	4.09
MnO	.11	tr. (?)	None	.71
S	—	5.47	5.46	—
SO_3	—	None	None	None
FeS_2	—	—	—	.49
P_2O_5	.13	.13	—	.20
BaO, SrO	.11	tr.	—	tr.
Cu	—	.25	Present	—
CO_2	.15	None	None	5.50
	99.93	102.00	98.15	101.01
Less O_2	—	2.05	—	—
Total	99.93	99.95	98.15	101.01

Analysts: W. F. Hillebrand and E. T. Allen.

1. Fresh unaltered Butte quartz monzonite.

2. Sericitized quartz monzonite, Parrot mine 1,300-foot level, 6 inches from vein.

3. Altered quartz monzonite, 1,000-foot level, Leonard mine, forms footwall of enargite vein.

4. Altered quartz monzonite, north crosscut from 700-foot level of Original mine (Propylite).

Analyses from Weed, W. H.: Geology and ore deposits of the Butte District, Montana. U. S. Geol. Surv. Prof. Paper 74, 1912, p. 88.

A word should be said at this point in regard to the character of the solutions responsible for the formation of tourmaline. The boron present in this mineral was very likely carried as boric acid, at least in the early stages of transport, but boric acid is a weak acid, and hence the solutions containing it would be alkaline according to our definition. Tourmaline, however, is generally accompanied by one or more of the fluorine minerals, in places in large quantities, which suggests that the net effect of the solutions that form tourmaline, is acid according to our definitions.

TABLE IV.

TOURMALINE ALTERATION.

	Rimini		Cactus		Braden		Caracoles	
	1	2	3	4	5	6	7	8
SiO_2	65.91	66.70	62.10	62.56	59.59	56.27	65.39	63.82
Al_2O_3	15.32	13.25	15.47	17.21	17.31	20.60	16.15	16.03
Fe_2O_3	2.28	1.34	2.64	2.29	3.33		1.08	None
FeO	2.02	.51	3.15	3.64	3.13		3.29	8.79
Total Iron					(4.78)	(4.54)		
MgO	1.52	.30	2.57	1.13	2.75	3.46	1.87	2.49
CaO	3.28	.05	5.31	.29	5.80	.49	3.07	0.37
Na_2O	3.08	.39	3.56	.07	3.58	(.30)*	2.28	None
K_2O	4.80	4.03	3.15	6.02	2.04	(4.40)*	5.35	3.74
H_2O-	.60	.08	.14	.14			0.11	
H_2O+	.60	.37	.72	2.70	1.26	(3.50)*	1.13	4.77
TiO_2	.59	.20	.81	.70	.77	.79	0.48	
CO_2	.21	.21	tr.	1.93				
P_2O_5	.18	.12	.27	.24	.26		.40	.36
MnO	tr.	tr.		.45	.18			tr.
BaO	.10	None						
Li_2O								
B_2O_3						(.70)*		tr.
F								
PbS		.59						
ZnS		1.86						
FeS_2		5.09						
FeAsS		4.75						
SO_3				.02				
S				.13		2.56		
Cu				.09		1.94		
Misc.						(.45)*		
	100.51	99.84	99.89	99.61	100.00	100.00	100.60	100.37

* Signifies that constituent has been estimated from mineralogical data.

1. Quartz-monzonite, fresh, Rimini, Mont. J. G. Fairchild, analyst.

2. Quartz-monzonite, altered, Rimini, Mont., hanging wall, Valley Forge mine. J. G. Fairchild, analyst.

3. Quartz-monzonite, fresh, Cactus mine. George Steiger, analyst.

4. Quartz-monzonite, altered, 2–4 inches from vein, Cactus mine. George Steiger, analyst.

5. Andesite, fresh (Daly's average andesite).

6. Andesite, altered, Braden mine. After F. E. Thackwell's partial analysis, recalculated to basis of 100 per cent and estimates made of other oxides from mineralogical data.

7. Fresh quartz monzonite, Caracoles, Bolivia. Helen Vassar, analyst.

8. Altered quartz monzonite, close to Lamero vein, Caracoles, Bolivia. Helen Vassar, analyst.

Analyses 1 and 2 from A. Knopf: The tourmaline silver-lead type of deposit. ECON. GEOL., VIII: 115, 1913.

Analyses 3 and 4 from B. S. Butler: *Op. cit.*, U. S. G. S. Prof. Paper 80, p. 76.

Analysis 5 from R. A. Daly: *Op. cit.*, p. 16.

Analysis 6 from F. E. Thackwell: Detailed statistical microscopic analyses of the ore and mill products of the Braden Copper Company, 1933. Unpublished Bulletin of the Microscopic Research Laboratories, Utah Engineering Experiment Station, Moscow, Idaho.

Analyses 7 and 8 from Lindgren, W.: Mineral deposits, 4th ed., p. 648. These are also in ECON. GEOL., XXI: 135–144, 1926.

Sericite.

Considerable attention has been given above to the general sericite problem, particularly as it concerns the removal or addition of potash. There it is shown that sericite alteration grades, on the one hand, into zones of complete replacement by silica and, on the other, to the propylite zones. The first of these is believed to represent acid attack, whereas the propylite zone, from its characteristic gain in bases, is believed to represent alkaline attack. Where then does the sericite zone stand?

Despite the gain in potash exhibited in sericite alterations, the net effect is a loss of alkalies and alkaline earths, as shown in Tables I–III; from two to five times as much of the other bases are lost as potash is gained. On general grounds, this points to the action of free acids; whether these are the strong acids, or merely CO_2, is not certain. It is well known that surface waters charged with carbon dioxide readily dissolve lime, soda, and magnesia (and potash with difficulty) from rocks, so there is little doubt that hydrothermal solutions charged with the same acid could effect the same alterations. Will potash, however, remain and form sericite in solutions of stronger acidity? This is the more immediate question in this discussion of the effects of strong acids on rocks. It is a well established fact that sericite is decomposed to kaolin by sulphuric acid in the supergene oxidation of sulphide bodies, and Day and Allen found that sulphuric acid had the same effect on sericite in the hot springs at Lassen Peak. They concluded that: "kaolin is a product of acid waters, whether hot or cold; where sericite is formed the waters were presumably alkaline." [24] Sericite, however, is well developed in greisens in association with fluorite, and with tourmaline in numerous tin deposits. In such occurrences, there is shown generally a slight loss in potash, or at the least, no notable gain. (See Table IV and p. 648 in Lindgren's "Mineral Deposits.")

In this connection, it is interesting to note the conclusions of

[24] Day, A. L. and Allen, E. T.: The volcanic activity and hot springs of Lassen Peak. Carnegie Inst., Wash., Pub. 360: 141, 1925.

Paige and Steiger [25] in regard to the influence of fluorine in the formation of sericite. It was found that the thoroughly and typically sericitized quartz monzonite in association with the copper deposits at Tyrone, New Mexico, contained a number of small veins of fluorite. An analysis of a representative specimen of the altered rock showed .09 per cent F., and it was believed that this was all contained in the sericite. From the proportions of sericite to the other minerals, the amount of fluorine in the sericite was calculated at 0.20 per cent. The presence of fluorine in mica is well established, so the fact that the sericite at Tyrone contains fluorine is not surprising, although the implications are rather suggestive of the stability of sericite in the presence of fluorine. Paige and Steiger believe that the hydrofluoric acid forms sericite from orthoclase by abstracting silica. The fluoride so formed is hydrolized to silica and the acid. The acid, according to them, thus acts like a catalyst, being used over and over again. This appears to be a reasonable reaction, but it does not account for the formation of sericite from the lime-soda fedspars. In such cases, potash must be added, as well as silica (and soda and lime) must be abstracted. Spurr [26] has also remarked upon the presence of fluorine in sericite, and has argued that this element is potent in the formation of the mica.

Potash, the critical component of sericite, is extremely tenacious in most geologic processes, a consideration that further suggests the stability of sericite under conditions of limited strong acid attack. In surface weathering, sericite as a recognizable mineral is destroyed, but potash (probably in some sub-microscopic equivalent of sericite) persists in almost the same amount in the resulting sediments as in the average igneous rock. Clarke [27] gives 3.13 per cent for the potash content of the average

[25] Paige, Sidney, and Steiger, George: Fluorine in sericitization. Wash. Acad. Sci. Jour., 8: 234–239, 1918. Paige, Sidney: Copper deposits of the Tyrone dist., New Mexico. U. S. Geol. Surv. Prof. Paper 122: 26–27, 1922.

[26] Spurr, J. E.: Geology of the Tonopah mining District, Nevada. U. S. Geol. Surv. Prof. Paper 42: 232–233, 1905.

[27] Clarke, F. W.: *Op. cit.*, p. 34. More than 5,000 analyses are represented in the igneous rock average, whereas the shale average represents but 78 analyses.

igneous rock, 2.80 per cent for the average sedimentary rock, and 3.24 per cent for the average shale. In contrast, there is 3.84 per cent of soda in the average igneous rock, 2.11 per cent in the sediments, and only 1.30 per cent in the average shale.

The foregoing evidences as a whole suggest that sericite as a mineral is not indicative of either alkaline or acid attack, but it probably *forms* (when potash is added) only from alkaline or weakly acid solutions. The sericite zones in hydrothermal alteration are, therefore, indicative of alkaline or faint acid attack only if they exhibit a distinct gain in potash. If potash remains constant, or decreases, even though sericite be present in abundance, intense acid attack (at one or another stage) is indicated. The evidence also suggests a possible difference between the behavior of sericite in solutions of the halogen salts and those of the sulphates.

Nacrite, Dickite and Kaolinite: Andalusite, Sillimanite, Topaz, etc.

The kaolin minerals are chemically midway between the mica-chlorite group and the alumino-silicates, andalusite, sillimanite, dumortierite, kyanite, topaz and zunyite. It is interesting, therefore, to note the recent trend towards the acceptance of nacrite and dickite as also allied to the mica and alumino-silicate groups genetically. In general, if not always, the presence of nacrite and dickite and the alumino-silicates (when they are of hydrothermal origin) is indicative of intense acid leaching. Bowditch [28] recently found this to be the case at Cerro de Pasco where dickite is associated with alunite and zunyite in the zones of intense silicification. According to Larsen,[29] the association of the clay minerals with alunite is a common one. Alunite contains the strong acid radical SO_3 and the formula of zunyite, according

It is thus probable that the figure for the potash content of the average shale is not as reliable as that in the average igneous rock, though it is not likely that the discrepancy is very great.

[28] Bowditch, S. I.: Ph.D. Thesis, Harvard University, 1935.

[29] Larsen, E. S., Jr.: Personal communication.

to Larsen and Berman,[30] is $Al_2O_3 \cdot SiO_2 \cdot Al(OH,FCl)_3$, which indicates 8.5 per cent of the halogens. Zunyite is thus much the same mineral as topaz, which contains 16–18 per cent F. in its formula of $2(AlF)O \cdot SiO_2$ (Larsen and Berman[31]), or $Al(F,OH)_2\ AlSiO_4$ (Ford[32]). The "kaolin" deposits of Cornwall (commonly associated with the tin lodes) are probably further examples of this double association of kaolin with strong acid minerals and acid leaching, though not all will agree that the kaolin in these deposits is of hydrothermal origin. Ross and Kerr,[33] for example, claim that the kaolin mineral is kaolinite and not either of the hydrothermal varieties of nacrite, or dickite. There certainly has been some surface weathering in the district, however, and it is possible that the material analyzed by Ross and Kerr was of this origin. (It seems probable to the writer, from the meager descriptions of these deposits available to him, that supergene as well as hypogene deposits exist within the district, which, if true, might account for the radical differences in opinion one finds in the literature on these deposits.) Lindgren[34] believes (from descriptions?) that the kaolin of the district formed from feldspar and sericite around sulphide bodies distinct from the tin lodes by the action of supergene sulphuric acid derived from the oxidation of the sulphides, yet he includes sketches showing the intimate association of kaolin with greisens alongside of tin veins. Davison[35] emphasizes the close areal association of the kaolin with the greisens and points out that, although the deepest workings on the main kaolin deposits are but 350 feet below the surface, none of them has been bottomed, and their tendency is to increase in size with depth. These evi-

[30] Larsen, E. S., Jr. and Berman, Harry: The microscopic determination of the nonopaque minerals. U. S. Geol. Surv. Bull. 848, p. 52.

[31] Larsen and Berman: *Op. cit.*, p. 113.

[32] Ford, W. E.: Dana's textbook of mineralogy. 4th ed., 1932, p. 613.

[33] Ross, C. S. and Kerr, P. F.: The kaolin minerals. U. S. Geol. Surv. Prof. Paper 165–E: 174, 1931.

[34] Lindgren, W.: Mineral deposits, p. 647.

[35] Davison, E. H.: Handbook of Cornish geology. Penzance, 1926, p. 60. See also Lilley, E. R.: The geology of some kaolins of western Europe. A. I. M. E. Tech. Paper 475: 14–20, 1932.

dences, if true, point to a hypogene origin of the deposits. It is also significant that the greisens, despite their content of sericite, show a loss in potash, a fact that suggests acid leaching of a type likely to give kaolin as an end product if carried far enough.

Another example of this same general type is the hydrothermal alteration of latite flows in the Frisco district of Utah [36] where andalusite is associated with kaolin (?), gypsum, fluorite, diaspore, specularite, and quartz in an igneous rock almost completely leached of potash, soda, lime and magnesia.

The andalusite-sillimanite lens at White Mountain, California is a further case of this type. The lens is believed by Kerr [37] to have developed through contact metamorphism and hydrothermal alteration of a trachyte, or an alumina-rich sediment. It adjoins an intrusive mass. Associated minerals are alunite, diaspore, tourmaline, topaz, sericite, and pyrophyllite. Both the structural and mineralogical features strongly suggest that the lens was formed by hydrothermal solutions.

Hydrothermal andalusite is said to occur in considerable quantities at the Boliden mine in Sweden in a sericitized zone bounding a core of sulphides, intense silicification, tourmalinization, and (collapse?) breccias. Further examples of hydrothermal andalusite, dumortierite ($8Al_2O_3 \cdot B_2O_5 \cdot 6SiO_2 \cdot H_2O$) and kyanite associated with tourmaline are found in North Carolina [38] and Nevada.[39]

The interrelationship of all of the examples listed in this subchapter is striking, and points to the same feature: leaching of alkalies and alkaline earths by strong acids; whether these acids were of direct magmatic origin or not is another question, but one which the writer is inclined to answer in the affirmative on the basis of the general arguments advanced throughout this paper.

[36] Butler, B. S.: Op. cit., Prof. Paper 80: 102, and 78–81.

[37] Kerr, P. F.: Andalusite and related minerals at White Mountain, California. ECON. GEOL., 27: 614–643, 1932.

[38] Stuckey, J. L.: Kyanite deposits of North Carolina. ECON. GEOL., 27: 661–674, 1932.

[39] Kerr, P. F. and Jenney, Phillip: The dumortierite-andalusite mineralization at Oreana, Nevada. ECON. GEOL., 30: 287–300, 1935.

Alumina and Silica.

In general, the "alumina constant" conception of Van Hise, Leith and Mead appears to hold in hydrothermal alteration except for the end stage of complete replacement by silica or sulphides. On the whole, silica is more stable than alumina, though it shows more fluctuations in the zones of less intense alteration. It is frequently added, whereas alumina is rarely introduced in recognizable amounts. The elimination of alumina from the zones of complete replacement by silica or sulphides is even more suggestive of acid attack than the removal of the other bases.[40] Alumina can be transported as a colloid, but otherwise only as salts of the strong acids, which facts place it on much the same basis as the sulphides. Soluble aluminates are known, but these are prepared in the laboratory only from caustic alkalis or through fusion, and are hence believed to be unlikely products of hydrothermal solutions.

It is stated above that alumina rarely appears to be introduced in rock alterations, and one may well wonder what becomes of that removed from zones of complete replacement. Perhaps alumina is commonly added to the neighboring rocks (as sericite) in amounts of the order of 1–3 per cent, which would be difficult to recognize as additions. It is probably also deposited as alunite in regions above the point of leaching. The Marysvale, Utah,[41] alunite veins probably represent such a case, for the alunite here appears to form largely as open-space filling, and there is certainly more alumina represented than could be reasonably ex-

[40] *Cf.* Lindgren, W.: The gold and silver veins of Silver City, De Lamar, and other mining districts in Idaho. U. S. G. S. 20th Ann. Rept. Pt. 3: 181, 1900. In speaking of the leaching of alumina, he says:

"... the loss of so much Al_2O_3 can be explained on the supposition that the waters contained sulphuric acid, as only such waters are known to dissolve alumina in such large quantities."

It is somewhat surprising that the significance of similar leaching of alumina elsewhere in replacement veins and in general silicification should have been subsequently so generally overlooked.

[41] Butler, B. S. and Gale, H. S.: Alunite—A newly discovered deposit near Marysvale, Utah. U. S. Geol. Surv. Bull. 511: 64, 1912. Callaghan, Eugene: Alunite deposits of the Marysvale region, Utah. Paper read before the Soc. Econ. Geol., New York, 1937. Abs. in ECON. GEOL., 32: 191, 1937.

pected to have come from the rocks through volume-for-volume replacement. The nature of the alteration in depth on these veins, or in the district, is unknown, but there is much silicification and leaching throughout the region occupied by the veins, so it is reasonable to suspect that there is at least some leaching in depth as well.

The transportation of silica is one of the great problems of ore geology, and so much has been said on the subject that the writer is hesitant to add anything further. The high vapor pressure of the halogen compounds of silica has been pointed out, and the implications of this fact were also discussed. The zoning of silicification is worth further emphasis, however, in this connection. Areas of complete silicification are always centrally located in relation to the channelways through which ore solutions are presumed to have travelled, and these zones are generally best developed towards the center and in the deeper portions of mineral districts. When rocks are completely replaced by silica it means that the relatively insoluble constituent alumina has been removed, as well as tenacious potash, and the more easily soluble bases. This is an obvious fact, but one of great significance to the acid-alkaline question if one grants that alumina and potash are not moved by hydrothermal solutions except when they are strongly acid.

Alunite, Anhydrite and Barite.

It generally is recognized that the sulphates as a rule, and especially alunite, favor the near surface environments.[42] This condition, according to the proponents of the alkaline view, points to two conclusions: first, the source of the acid radicals is not magmatic, for if it were the free acid would have been neutralized during the course of the supposedly long transit from source to surface, second, the acid is produced from alkaline solutions through the interplay of low pressures (near surface) and high temperatures[43] (because of residual heat in the host rock channel,

[42] Lindgren, W.: Mineral deposits, p. 458. Butler, B. S.: *Op. cit.*, ECON. GEOL., 14: 1919.

[43] Graton, L. C. and Bowditch, S. I.: Alkaline and acid solutions in hypogene zoning at Cerro de Pasco. ECON. GEOL., 31: 651–698, 1936.

or other causes), or by direct surface oxidation of H_2S or S in the solutions. Butler [44] has suggested also that either or both H_2S and S may oxidize to SO_3 through contact with ferric minerals during passage from source to outlet.

In his excellent summary of the occurrences of the sulphate minerals, Butler [45] cautiously suggests the possibility that in some cases the sulphuric acid responsible for their information may be direct magmatic in origin, but this view is subordinated in space and emphasis to a discussion of the schemes for meta-magmatic production of the acid, since to him the sulphates did not appear near-source in position.

As an indication of a gradual shifting of ground towards the direct magmatic viewpoint, it is of interest to note the recent opinions of Burbank [46] expressed in his report on the ore deposits at Bonanza, Colorado. He believes that the solutions which accomplished the extensive silicification and alunitization of the local rock masses were of direct-magmatic or primary acidity. The ore deposits, however, he believes are of later age and formed by alkaline solutions. The presence of sericite in the portions of the generally silicified wall rocks immediately adjacent to the veins in his evidence of the later age and alkaline nature of the solutions that deposited the ores.

In regard to the general impression that sulphates favor near-surface environments, it is significant that several deposits have been found to increase in quantity in depth, but two others have been encountered only below the surface. Such occurrences are not believed to indicate that the sulphates increase downward in the depth-zone scale of ore deposits as a whole, for this is surely not the case. To the writer, however, the persistence and increase of the sulphates in depth in some of the better explored occurrences does suggest that in these deposits they are near-source as well as near-surface in position.

Anhydrite.—In regard to the increase of sulphates in depth in

[44] Butler, B. S.: *Op. cit.*

[45] Butler, B. S.: *Op. cit.*

[46] Burbank, W. S.: Geology and ore deposits of the Bonanza mining district, Colorado. U. S. Geol. Surv. Prof. Paper 169: 82–85, 1932.

individual deposits, the occurrence at Morococha, Peru, is particularly instructive. Here the sulphate mineral anhydrite is practically unknown above the 700 foot level, but below this horizon the limestones of the district are extensively replaced by it, and on each successively lower level explored the total quantity increases greatly. The indications from the present lowest level 1,600 feet below the surface point to much greater quantities of anhydrite on this level than on any of those above.

Somewhat the same is the case at Britannia Beach, British Columbia. The ore bodies here consist of five contiguous pyrite replacement deposits formed along a strong shear zone in an island or large roof-pendant of volcanics in the Coast Range Batholith. Small amounts of chalcopyrite with gold give the deposits economic value. The alteration in the walls adjacent to the ores is described as varying from chlorite-sericite to complete silicification, with the latter dominant. According to James,[47] "anhydrite is very abundant" as discrete veins in the altered rock and occasionally in the ores. These are from 1–10 feet in width, and increase in number and total quantity in depth. In the Fairview mine extensive sulphide mineralization of the "hanging-wall zone" above the 600 level gives way on the 850 and 1000 levels to replacement veinlets of anhydrite. The total amount of anhydrite represented in the area must be of the order of several million tons, and this at depths of 2,500 feet below the present surface, which must lie at least 5,000 feet below that obtaining at the time of ore deposition. It is of further interest to note again the association of intense silicification with the development of acid minerals, in this case a sulphate.

At the Braden mine in Chile enormous quantities of anhydrite occur as 1–10 per cent disseminations through andesites in association with pyrite-chalcopyrite mineralization, and introduced biotite and tourmaline with local areas of intense silicification. This occurrence will be described in a forthcoming paper, but the point is made now that the amount of anhydrite appears to increase with depth, and is recognized only below the 1000-level.

[47] James, H. T.: Britannia Beach map-area, British Columbia. Can. Geol. Surv. Mem. 158, 1929.

This is probably a consequence of extensive leaching in the upper portions of the mine by surface waters, but at any rate, there is no diminution in quantity in the 1000-foot range below this zone of leaching.

Alunite.—In the East Tintic area of Utah,[48] the sulphate mineral alunite is associated with intense silicification that grades outwards and upwards to sericite and propylite types of alteration. The central Big Hill stock of quartz porphyry is "drenched" with alunite and silica, and one finds in the deeper and central portions of the "ore foci" of the Tintic Standard and other mines that the same zoning obtains.

The iron deposits of South Anhui, China, according to Hsieh [49] are associated with abundant alunite and kaolin, which he takes to represent the action of solutions rich in direct magmatic sulphuric acid. It is interesting to note that the sequence in these large replacement bodies is magnetite with apatite and actinolite followed by hematite, alunite and kaolin. In other words, the development of the sulphate was late, and accompanied by oxidation of the pre-existing iron oxide, which is just the reverse of the process favored by Butler. Some three hundred miles to the southeast at Ping Yang there occur enormous deposits of alunite (one billion tons estimated reserves) as selective replacements of certain volcanic flows. Yih [50] favors the idea that the source of the responsible acid is in the monzonite stocks that intrude the volcanics. Similar deposits of alunite, commonly with pyrophyllite are widely distributed in this section of China.

A deposit of alunite and pyrophyllite of large magnitude is also described from British Columbia.[51] This occurrence lies in the midst (7,500 feet stratigraphically from the top) of a 10,000-foot series of andesite and dacite flows and fragmentals within

[48] Billingsley, Paul: Titanic mining district in Guidebook 17, XVI Int. Geol. Cong., pp. 108–124, 1933.

[49] Hsieh, C. Y.: The iron deposits of South Anhui. Bull. Geol. Soc. of China, 10 (Grabau Anniversary Number): 317–351, 1931.

[50] Yih, L. F.: The alunitization and pyrophyllitization of the rhyolite and tuff in some maritime districts of South Eastern China. Nat. Res. Inst. Geol., Mem. XI, Shanghai, 1931.

[51] Clapp, C. H.: Alunite and pyrophyllite in triassic and jurassic volcanics at Kyuquot Sound, British Columbia. ECON. GEOL., 10: 70–88, 1915.

two or three miles of an off-shoot of the Coast Range Batholith. To judge from numerous small intrusives in the volcanic pile, it seems likely that this intrusive mass probably underlies the area at comparatively shallow depths. Clapp, however, believes that the alunitization of the volcanics is unrelated to the intrusive, having been effected much earlier during an intra-volcanic period. This he believes indicated by the opal-like nature of the silica, which is taken as an indication of near-surface formation. He also believes that the weight of opinion is against the idea that the source of the acid could have been magmatic, so rejects the possibility. The altered rocks as a whole show a low degree of porosity (Table V) considering their extensive re-working, and it seems that this fact may indicate that the replacement was effected under considerable pressure, such as would obtain in a

TABLE V.

ALUNITE AND PYROPHYLLITE ALTERATION, KYUQUOT SOUND, B. C.

	1	2	3	4	5	6
SiO_2	73.22	87.80	81.94	71.88	48.82	62.70
Al_2O_3	13.46	9.08	15.29	23.56	19.08	12.68
Fe_2O_3	2.33	.40	.11	.14	.07	1.40
FeO	.96	n.d.	n.d.	tr.	n.d.	tr.
FeS_2	—	—	—	—	—	2.69
MgO	.42	n.d.	n.d.	.21	n.d.	.05
CaO	1.50	n.d.	n.d.	.06	n.d.	.20
Na_2O	5.46	.02	.40	.36	2.74	1.09
K_2O	1.74	1.70	.50	.43	4.40	2.10
H_2O	.62	1.04	2.40	3.24	7.00	7.15
TiO_2	.28	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	.10	n.d.	n.d.	n.d.	n.d.	n.d.
MnO	tr.	n.d.	n.d.	n.d.	n.d.	n.d.
SO_3	tr.	tr.	tr.	tr.	17.32*	7.06*
S	—	—	—	—	.57*	2.88*
Total	100.09	100.04	100.64	99.88	100.00	100.00
Per cent porosity	.67	1.8	1.2	1.0	2.1	1.8

* S and SO_3 were not determined directly. Both were estimated together as SO_3 and relative amounts of S and SO_3 arrived at by calculation.

All analyses by N. L. Turner, Dept. of Mines, Ottawa, Canada.

1. Dacite tuff.
2. Cherty quartz-sericite rock.
3. Quartz pyrophyllite rock (pink).
4. Quartz pyrophyllite rock (gray).
5. Quartz-alunite rock (pink). 45 per cent alunite.
6. Quartz-alunite rock (gray). 18 per cent alunite.

Reference: Clapp, C. H.: ECON. GEOL., X: 1915.

7,500-foot pile of flow rocks. The proximity to the large intrusive mass likewise suggests that the mineralization is post-volcanic and related to the intrusive. It is significant in this connection that the rather similar occurrence of sulphates (anhydrite) and silification at nearby Britannia Beach is directly related to this batholith.

The Cerro de Pasco occurrence of alunite is insignificant in total quantity when compared with any of the foregoing examples. The mineral is rarely present in amounts greater than 5 per cent, and is generally seen only in microscopic preparations. The distribution of the mineral is equally limited, being confined,

TABLE VI.

KAOLINIZATION, SILICIFICATION, BONANZA, COLORADO.

	1	2	3	4	5	6	7	8	9
SiO_2	57.62	59.66	54.23	88.73	61.28	66.48	95.18	93.17	82.57
Al_2O_3	16.84	16.09	18.82	.97	2.42	1.64	1.20	2.52	11.44
Fe_2O_3	3.05	2.57	1.69	3.50	31.49	.64	.77	.19	.16
FeO	3.9	3.53	4.06	3.56	2.49	2.74	2.27	1.92	.65
MgO	2.14	2.29	2.25	.05	.05	tr.	n.d.	.03	tr.?
CaO	4.81	4.48	6.62	.24	.28	.20	n.d.	.05	00
Na_2O	3.07	3.08	3.91	n.d.	tr.	tr.	tr.	tr.?	tr.?
K_2O	4.95	4.92	3.08	n.d.	.19	.27	tr.	.15	1.89
H_2O-	.24	.25	.08	.02	.05	tr.	00	.09	.43
H_2O+	1.39	1.84	1.51	.69	.58	1.64	.40	1.03	2.98
TiO_2	1.30	1.00	1.43	1.50	.50	1.45	.25	.63	.35
CO_2	1.06	.10	2.13	n.d.	.00	n.d.	n.d.	—	—
P_2O_5	.44	.35	.16	.20	.20	.43	.06	.10	.09
SO_3	.05	—	tr.	n.d.	.28	.00	tr.	.20	1.2
Cl	n.d.	—	tr.	n.d.	tr.	.09	n.d.	—	—
F	n.d.	—	—	n.d.	.00	.00	n.d.	.00	.00
S	n.d.	—	tr.	n.d.	.06	Present	n.d.	—	—
MnO	.15	—	.17	n.d.	n.d.	n.d.	n.d.	—	—
BaO	.04	—	—	.00	.30	tr.?	.00	.07	.06
TiO_2	n.d.	—	tr.	—	—	—	—	—	—
FeS_2	n.d.	—	—	—	tr.	24.96	—	—	—
ZnS	n.d.	—	—	—	.00	.05	—	—	—
Total	100.05	100.16	100.14	99.46	100.17	100.59	100.13	100.15	100.74
Porosity	0.7%	1.3%	—	1.1%	5.1%	3.3%	3.4%	7.5%	—

1. Fresh augite-mica andesite.
2. Fresh gray quartz latite.
3. Augite andesite with secondary calcite, chlorite and sericite.
4. Altered andesite of red jasper type.
5. Altered andesite of black jasper with hematite (32 per cent).
6. Altered andesite of red jasper with pyrite (25 per cent).
7. Silicified andesite (95 per cent) quartz.
8. Silicified volcanic rock with sericite kaolin (0.15 K_2O).
9. Silicified volcanic rock with kaolin minerals (1.89 K_2O).

Reference: Burbank, W. S.: U. S. Geol. Surv. Prof. Paper 169, p. 78.

Analyses 1, 2, 4, 5, 6 and 7: by S. G. Fairchild. Analysis 3: by George Rohmer and E. V. Titus. Analyses 8, 9: by (?). Porosity calculated by O. C. Schmedeman from Burbank's data on specific gravity.

TABLE VII.

ALUNITIZATION AT ROSITA HILLS, COLORADO.

	1	2	3	4
SiO_2	70.87	65.94	69.67	76.22
TiO_2	tr.	—	—	0.11
Al_2O_3	15.18	12.95	13.72	19.45
Fe_2O_3	2.18	.33	—	tr.
FeO	0.12	.07	—	—
MnO	tr.	tr.	—	—
BaO	—	tr.	—	—
CaO	1.58	.10	.07	tr.
MgO	0.60	.05	tr.	tr.
K_2O	5.04	2.32	2.44	tr.
Na_2O	3.47	1.19	.34	tr.
H_2O	1.08	4.47	4.73	3.82
SO_3	—	12.47	9.27	.29
P_2O_5	tr.	—	—	.13
	100.12	99.89	100.24	100.02

Analysis by L. G. Eakins.

1. Fresh rhyolite from Pennsylvania Hill.
2. Rhyolite altered to quartz and alunite (32 per cent). Democrat Hill.
3. Rhyolite altered to quartz and alunite (24 per cent). Mount Robinson.
4. Rhyolite altered to quartz and diaspore. Mount Robinson.

Reference: Cross and Emmons. U. S. Geol. Surv. 17th Ann. Rept. Pt. 2, pp. 315–317 and 324.

TABLE VIII.

ALUNITIZATION AT GOLDFIELD, NEVADA.

	1	2	3	4
SiO_2	60.86	60.75	160.07	151.27
Al_2O_3	16.01	15.37	42.11	38.27
Fe_2O_3	3.39	.20	8.91	.50
FeO	2.37	.14	6.23	.35
FeS_2	None	7.23	None	18.00
MgO	2.77	.06	7.28	.15
CaO	5.92	.41	15.57	1.02
Na_2O	3.12	.84	8.21	2.09
K_2O	2.56	1.06	6.73	2.64
H_2O-	.97	1.33	2.55	3.31
H_2O+	2.03	6.62	5.34	16.49
SO_3	None	5.99	None	14.91
	100%	100%	263.00	249.00

Analyses by George Steiger.

1. Fresh dacite: recalculated to summation of 100, with minor constituents omitted.
2. Altered dacite: recalculated to summation of 100, with minor constituents omitted.
3. Constituents, in grams, in 100 c.c. of fresh dacite with 0.9 per cent porosity.
4. Constituents, in grams, in 100 c.c. of altered dacite with 9.9 per cent porosity.

Reference: Ransome, F. L.: U. S. Geol. Surv. Prof. Paper 66, p. 181.

according to Bowditch,[52] to the silicified sheaths alongside of the copper-bearing veins. Bowditch cites the fact that there is no noticeable increase of the sulphate on the lowest level as an indication that it does not represent attack by magmatic free acids. He and Graton [53] reason that if it were of deep-seated origin, the proportion should increase with depth. It is true that there is no noticeable increase in the average content of alunite in the thin sections that have been made of the vein zones on the 2,100 level, but it is questionable whether the total quantity of alunite on the level as a whole does not show an increase over that obtaining on the levels above. It is significant in this regard that the only macroscopic occurrences known in the district to date are on the 2,100 level, and two of these contain a few hundred tons each. The level is only partially developed, so it is impossible to judge what the total amount of such concentrations may be, but it is not unlikely that this level may prove to contain considerably more alunite than any of those above it.

Barite.—In the foregoing, little has been said of the occurrences and significance of barite. This mineral, although of widespread distribution in ore deposits, rarely occurs in impressive concentrations, such as do anhydrite and alunite (probably because barium is a minor constituent of rocks). For this reason, it is difficult to single out especially illustrative or critical examples. Perhaps the most significant feature about its occurrence is its ubiquity. This may be taken as an indication of the frequency of sulphuric acid in ore deposits, which is made evident by the relative insolubility of the mineral. It seems that many of the occurrences of barite are much too deep-seated to be explained by any process dependent upon surface oxygen, or the Graton-Bowditch low pressure-high temperature reaction. The numerous pyrite lenses in schists, such as those of Shasta County, California [54] (which also contains anhydrite), and Kyshtim,

[52] Bowditch, Samuel I.: Ph.D. Thesis, Harvard University, 1935.

[53] Graton, L. C. and Bowditch, S. E.: *Op. cit.*

[54] Graton, L.C.: The occurrence of copper in Shasta County, California. U. S. Geol. Surv. Bull. 430: 71–111, 1910.

Russia,[55] are examples of the point. It is also of interest to notice the rather common association of barite with fluorite.[56] In such cases, the deposits are often thought to have formed relatively near the surface, but it appears likely that this is not always a necessary conclusion as instanced by the deposits of the Appalachians described by Stuckey and Davis.[57]

Anhydrite and Alunite.—Anhydrite and alunite are much more soluble than barite, the sulphides, or most of the rock minerals, and it is perhaps because of this fact that these two sulphate minerals are rather uncommon in ore deposits, particularly in the deep-seated bodies formed at high temperatures and pressures. Near-surface environments are the usual habitat of these minerals. This may be due to the physical nature of these positions, *i.e.*, general low-pressure conditions, and numerous fractures that allow rapid dissipation of heat. When such sites are also near-source in position, they would be theoretically favored for the formation of sulphates in abundance (if the source of the acid is magmatic).

In the volcanic vents at Llallagua, Braden, and Potosi anhydrite and alunite are associated with notable silicification and leaching, and with minerals generally believed to form at high temperatures. It is possible that the requisite high temperatures were furnished in these near-source localities from the residual heat of intrusives within or around the vents, or from the passage of large volumes of ore solutions. However, when the presence of these rather anomalous high temperature minerals is taken into account, together with that of the abundant sulphates and general acid attack, the most direct explanation is that these particular vent deposits were formed close to the source. Nearness to the point of emanation of the solutions thus accounts for both features of these deposits, *i.e.*, their acid and high temperature nature is the result of primary heat and acidity of the solutions,

[55] Stickney, A. W.: The pyritic copper deposits of Kyshtim, Russia. Econ. Geol., 10: 594–633, 1915.

[56] Lilley, E. R.: The economic geology of mineral deposits. Holt, New York, 1936, pp. 724–738. Lindgren, W.: *Op. cit.*, pp. 398–402.

[57] Stuckey, J. L. and Davis, H. T.: Barite deposits in North Carolina. Tr. A. I. M. E., 115: 346–355, 1935.

and the reason these qualities prevailed without dissipation or neutralization up to surficial horizons is because the origin of the solutions was likewise near-surface. Areas of Tertiary volcanic outbreak are the common locale of the sulphate minerals, and it is possible that many of these sites were likewise near-source in nature at the time the volatile products of their source magma formed ore and sulphate deposits in them.

CONCLUSIONS.

In conclusion, the question of the "acid" minerals in ore deposits is tied to two fundamental questions of far-reaching nature. One of these is the general problem of the nature of ore solutions as they leave the magma source, and as they ascend and deposit their loads. That these solutions may be acid at the start is now accepted by many; that they continue acid to the point at which known ore deposits have been formed is the main contention of this paper, although it is admitted by few geologists. The other fundamental question is directly tied to this problem of how far from the magma ore solutions may remain acid. In simplest terms, it involves a decision on whether or not near-surface deposits such as those commonly found in volcanic flows and vents may also be near-source in position. If one assumes that the ore solutions start their journeys in an acid state, then it follows logically that where acid minerals and acid leaching are developed in abundance those points must be comparatively near-source. If such logic be admitted, the vent and volcanic deposits as a group are near-source. If one does not agree that the solutions are originally acid, then it does not follow that acid minerals and leaching are indicative of near-source positions. The problem then is to account for the sulphates on some scheme based on the preferred near-surface occurrence of these minerals, and the proponents of such schemes may well deny that vents and volcanic regions are near-source at the time ores form in them. This is a question of opinion that can be solved only by recourse to general arguments and the usual circuitous reasoning of geologic problems.

The burden of proof is admittedly on those who favor the concept that ore solutions are originally acid and that they continue so to points within the limits of observation. To prove their point, they must test and challenge the results of fifty years of observation and thinking based on wholly different ideas, and they must substitute a complete theory to account for all of the many and varied features of ore deposits if their premise is to gain general acceptance. The field is tremendous and many difficult problems must be met, but the rewards may be proportionately great.

This paper has but scratched the surface of the field, though it indicates vital inconsistencies in the prevailing views, and suggests that they might be reconciled by fuller consideration of the probability that the metals leave the magma as halides in the gaseous state, and are transported in many or all cases in the liquid state as chlorides, fluorides and sulphates to and above the lower limits of our range of observation.

OROYA, PERU, S. A.,
Aug. 25, 1938.

MAGNETITE CRYSTALS FROM METEORIC SOLUTIONS.

KIRIL SPIROFF.

ABSTRACT.

Primary magnetite crystals occur in vugs and as a filling in leached iron-ore conglomerate found on the Mesabi Range, Minnesota. The associated minerals are marcasite, goethite, hematite, and siderite. The host material is a porous mechanical concentrate of the weathered underlying iron formation. It shows leaching, probably due to the activity of sulphate waters. Chemical reactions given indicate the possibilities that the magnetite is a precipitate from meteoric waters. Crystals grown in the laboratory under low pressure and temperature conditions are magnetic, and X-rays reveal the same diffraction pattern as that of magnetite from other sources. From the field investigation and from magnetite crystals developed in the laboratory the conclusion is drawn that magnetite can be formed as a precipitate from meteoric waters.

INTRODUCTION.

On the Mesabi Range[1] at the Kinney Mine, Kinney, Minnesota, primary magnetite crystals are found in vugs and as filling between limonite and leached iron-formation pebbles in an iron-ore conglomerate. This unusual occurrence of magnetite has been described and discussed.[2] Recently, the writer made further study of the magnetite. He describes here briefly the associated minerals and the results of his field observations and laboratory experiments, and states why he believes the magnetite to be a product of the activity of meteoric waters.

GENERAL GEOLOGY OF THE DISTRICT.

Reviewed briefly, the general geologic conditions on the Mesabi Range, west of Virginia, Minnesota, are as follows:

The Middle and Upper Huronian gently-dipping sediments,

[1] Magnetite crystals in conglomerate vugs are also found at the Brunt Mine, and as filling in the conglomerate at the Scranton and Alpena Mines.

[2] Gruner, J. W.: Magnetite cementing certain ore conglomerates on the Mesabi Range. Econ. Geol. 29: 757, 1934.

with monoclines, terraces, and domes, were somewhat folded and faulted before they were subjected to weathering and erosion. Then, according to the record revealed by the sedimentary succession, came the deposition of iron-ore conglomerate (possibly Cambrian) and of sediments of Cretaceous and probably of later ages. Lastly, the topography was considerably modified by glaciation, which left a mantle of glacial debris covering remnants of the various iron-ore conglomerates and shales. In many of the mines this succession can be seen clearly, or is indicated by the information available.

J. F. Wolff of the Oliver Iron Mining Co. believes that there are two different conglomerates overlying the iron formation. The writer contends that there are three. In re-examining with J. F. Wolff some of the conglomerate outcrops, superimposed conglomerates were seen. The evidence that these may yield for determining the ages specifically has not been worked out as yet. Much material has been gathered, and it is hoped that in the near future the problem may be properly dealt with.

THE MAGNETITE.

The magnetite crystals are octahedrons, some of which are as large as one inch in diameter. Few are modified by dodecahedron faces. They show good cleavage, have a black streak, and are magnetic. Polarity has not been observed. Most of the crystals in the vugs are clean and bright; a few, however, are coated with an amorphous, whitish substance, a hydrated ferric sulphate; others show coatings of "limonite"; and some have crystals of marcasite and siderite upon them.

A chemical analysis of the magnetite crystals, made by R. F. Makens, shows 71.74 per cent total iron with a trace of insoluble. This indicates that nearly all of the iron is in the form of ferrous-ferric oxide (Fe_3O_4) and that very little ferric oxide (Fe_2O_3) is present, a conclusion further substantiated by a study of polished specimens.

Powdered specimens of the Kinney Mine magnetite were placed in one end of a capillary tube; and powdered magnetite

crystals from the Champion Mine, Michigan, were placed on the other end. Diffraction patterns[3] were obtained on a General Electric Unit. The interplanar spacings and the crystal symmetry of the Kinney Mine magnetite specimen were shown to be identical with those of the Champion Mine magnetite.

Iron sulphide occurs with the magnetite crystals. Most of this sulphide is in botryoidal or spherical masses whose cross-sections show radial structure and whose outer bounding surfaces have rectangular crystal faces—conditions suggesting marcasite. Polished specimens under polarized light show the mineral to be anisotropic and hence marcasite. However, not all of the iron sulphide in the conglomerate is marcasite; there are a few small isolated pyrite crystals in the form of octahedrons modified by cubes.

Flat rhombohedrons of the calcite group are common. Occasionally, white, resinous, barrel-like crystals of an unidentified mineral are found. These are hexagonal, with prismatic and basal habit. They have a perfect basal cleavage and a prism cleavage that varies from distinct to poor.

Thin sections reveal this unknown mineral to be uniaxial positive (+) with high birefringence. Indices are $\alpha = 1.570\pm$, $\beta = 1.575\pm$, $\gamma = 1.598\pm$.

Spectographic analysis[4] shows the presence of silicon, magnesium, titanium, aluminum, calcium, and iron.

GENERAL DESCRIPTION OF THE CONGLOMERATE.

The host material of the magnetite is a conglomerate with well-rounded pebbles, some of hematite and limonite, and others of decomposed taconite that shows bedding. The individual layers of the bedded pebbles consist of dense hematite or limonite, porous hematite and limonite, and decomposed taconite. The larger pebbles of the conglomerate are somewhat angular, but the corners are well rounded. The matrix consists of smaller grains of the same materials as the pebbles—that is, of the un-

[3] R. W. Drier.
[4] R. F. Makens.

derlying iron formation. All of the material shows that it underwent leaching as well as cementation after the conglomerate was deposited.

In the large fractures and vugs of the conglomerate at the Kinney and Brunt Mines, some of the cementing material has crystallized into magnetite and marcasite. These are the striking and outstanding cements, but they are not the most abundant. The common iron cement is composed of iron oxide and hydroxide in the form of goethite and hematite. The common crystals in the vugs are members of the calcite group—siderite, dolomite, and calcite.

Age of the Conglomerate.—At the Kinney Mine this conglomerate lies unconformably on top of the weathered iron formation. It is found in a flat U-shaped valley that suggests a filled river channel, and which crosses the iron formation nearly at right angles to the trend of the ore-bearing trough. When the conglomerate was excavated, F. H. Cash, Superintendent of the Kinney Mine, found some fossil wood but did not observe other fossils. Chemical analyses made at the Kinney Mine Laboratory and released through the courtesy of Mr. Cash are as follows:

	Iron	Phos.	SiO_2	Mn	Al_2O_3	CaO	MgO	S	Loss on Ignition
No. 1.....	49.94	.067	11.22	.80	8.04	.26	.61	.072	7.18
No. 2.....	50.88	.067	11.10	.61	8.33	.21	.60	.213	5.80

The material used in analysis No. 2 consisted chiefly of large well-cemented blocks, of which many contained more magnetite crystals, and also more sulphur, than the material used in analysis No. 1. The alumina content of both groups is relatively high, much higher than that of most members of the iron formation.[5]

At the Brunt Mine the conglomerate unconformably overlies weathered iron formation, which is flat and limonitic, having many cavities filled with siderite crystals. The appearance of the conglomerate varies from place to place, depending upon the material from which the sediments were derived and upon the

[5] Paint rock member has a high alumina content.

conditions to which the conglomerate has been subjected. The hardness of the pebbles is variable; those cemented by marcasite and magnetite are relatively soft and porous, whereas those cemented by limonite are much harder. The hardest ones are the uncemented, polished, water-worn pebbles.

A smooth, water-worn pebble of limonite found in the Bennett Mine at the bottom of the Cretaceous sediments was placed in a beaker containing dilute sulphuric acid. After a month—a period during which the solution evaporated considerably—the pebble, as well as the bottom of the beaker, was covered with white encrustations. On the pebble, underneath this deposit, was a porous zone of fine, deep-red, earthy hematite. This condition is typical of the pebbles in the conglomerate at the Kinney, Scranton, and Brunt Mines, but has not been observed at the Bennett Mine. A possible explanation of the character of the Kinney Mine pebbles is that a chemical reaction of the above nature, involving the activity of sulphate waters, took place.

The material containing the magnetite crystals is composed of fragments of the iron formation and shows considerable selective concentration; in places it warrants shipment as iron ore. This fact perhaps demonstrates that the conglomerate is a mechanical concentrate of the weathered iron formation.

ORIGIN OF THE MAGNETITE.

Two possible explanations of the origin of the magnetite have been offered: first, that it was formed by deposition from meteoric solutions; second, that it was developed by magmatic or hydrothermal processes.[6]

The occurrence of magnetite as a product of meteoric deposition, although apparently unusual, is not unknown. H. Rosler[7] observed magnetite in clay deposits from rivers and seas. C. W. Von Gumbel[8] observed magnetite in freshly formed sediment at

[6] Hydrothermal is here used to describe waters wholly or partly of magmatic origin.

[7] Rosler, H.: Neues Jahrb. Min. 15: 231, 1902.

[8] Von Gumbel, C. W.: Sitzber. Bayr. Akad. 15: 417, 1886.

the bottom of the sea. Magnetite cores [9] are found in some of the hematite pisolites or granules that occur, after serpentine, in the upper part of the Cuban residual iron ores, where the magnetite is most certainly a result of precipitation during weathering.

Magmatic and hydrothermal occurrences of magnetite are so common that they lead to the assumption that all magnetite [10] is formed under conditions of high temperature. Temperature is an important factor in determining the stability of minerals, but it is not the only one.

In this paper the writer supports the theory that the magnetite was deposited by supergene solutions, and presents arguments based upon the chemistry of the processes under discussion, upon laboratory experiments, and upon field observations.

CHEMISTRY AND DEPOSITION.

In studies of circulating meteoric waters in sediments containing gypsum, and hydrocarbons,[11] it has been noted that the gypsum is reduced by the hydrocarbons to form CaS, which when combined with water yields hydrogen sulphide. The liberated hydrogen sulphide generally reacts with iron salts to form marcasite.

The formation of marcasite under other conditions is well known, both from field observations and from laboratory experiments.[12]

[9] Leith, C. K., and Mead, W. J.: Origin of the iron ores of Central and Northeastern Cuba. A. I. M. E. Trans. XLII: 90–102, 1911.

[10] Gruner, J. W.: Additional notes on secondary concentration of Lake Superior Ores. ECON. GEOL. 27: 189–205, 1932.

[11] Mellor, J. W.: A comprehensive treatise on inorganic and theoretical chemistry, 10: 122.

[12] "The formation of iron disulphide was accomplished (1) by the action of hydrogen sulphide on ferric salts, or the action of hydrogen sulphide and sulphur on ferrous salts; (2) by the addition of sulphur from solution to amorphous ferrous sulphide or pyrrhotite; (3) by the action of soluble polysulphides on ferrous salts; (4) by the action of soluble thiosulphates on ferrous salts."—Concerning the action of hydrogen sulphide on ferric salts, the writers say, "The first action of hydrogen sulphide on ferric salts is, of course, the immediate reduction of the latter and the simultaneous precipitation of sulphur:

$$(1)\ Fe_2(SO_4)_3 + H_2S = 2\ FeSO_4 + S.$$

Observation of marcasite [13] shows that it has a pronounced tendency to decompose in the presence of oxygen and moisture.[14] Although the chief agent is oxygen in fresh water, carbonic acid [15] contained in meteoric waters is also commonly active in decomposing marcasite.

It is well known that in many places iron oxides and hydroxides are precipitates from ferruginous waters, the so-called chalybeate waters. Such waters transport the iron, probably as a ferrous hydrocarbonate, and normally yield precipitates of goethite, and hematite. The reactions involved may be expressed by the following equation:

$$(5)\quad 2\ Fe(HCO_3)_2 + 4\ H_2O = 2\ Fe(OH)_2 + 4\ H_2CO_3.$$

However, ferrous hydroxide is not stable and rapidly oxidizes as soon as the carbonic acid is removed or in the presence of oxygen-bearing waters. The latter reaction may be represented by the following equation:

$$(6)\quad 2\ Fe(OH)_2 + O = Fe_2O_3 + 2\ H_2O.$$

If conditions were such that the chalybeate waters came in contact with the solutions that were forming the marcasite, the following might be expected:

(*A*) The formation of the marcasite would be terminated. In fact some decomposition of previously formed marcasite [16] might take place.

In a closed vessel where the hydrogen sulphide is prevented from oxidation or escape, a secondary reaction proceeds, viz.:

$$(2)\quad FeSO_4 + S + H_2S = FeS_2 + H_2SO_4.$$

Allen, E. T., Crenshaw, J. L., and Johnson, John: The mineral sulphides of iron. Am. Jour. Sci. ser. 33, pp. 169–236, 1912.

[13] On the protected spots of the pit bank, white fibrous encrustations of melanterite can be seen.

[14] "The oxidation of the sulphide is started by oxygen and hastened by the ferric sulphate developed. At the surface it proceeds as follows:

$$(3)\quad FES_2 + 7O + H_2O = FeSO_4 + H_2SO_4,$$

$$(4)\quad 2\ FeS_2 + 15O + H_2O = Fe_2(SO_4)_3 + H_2SO_4.$$

This last reaction involves several intermediate stages during which ferrous sulphate, sulphur dioxide, sulphur, or hydrogen sulphide may form." Lindgren, Mineral deposits, 3rd ed., pp. 936, 1928.

[15] Beck, R.: The nature of ore deposits, 1st ed. 2: 369.

[16] "Oxidation of ferrous to ferric iron is retarded by free acid." Locke. Leached outcrops as guides to copper ore, p. 37.

(*B*) In turn, the presence of the marcasite-forming solutions would prevent the chalybeate waters from forming the normal ferric oxide [17] or hydroxide precipitates.

(*C*) (i) As the solutions mingle, the mixture of ferrous and ferric salts could be expressed by a combination of equations (1) and (5), and the resulting product would be entirely different from the one formed when each solution precipitated separately. The composite reaction may be expressed by the following equation:

$$(7)\quad Fe_2(SO_4)_3 + H_2S + Fe(HCO_3)_2 + 4\ H_2O = Fe_3O_4 + 4\ H_2SO_4 + 2\ H_2CO_3.$$

(ii) Or, if equations (4) and (5) are used in a similar manner, the reactions may be expressed by the following equation:

$$(8)\quad Fe_2(SO_4)_3 + Fe(HCO_3)_2 + 4\ H_2O = Fe_3O_4 + 3\ H_2SO_4 + 2\ H_2CO_3.$$

Equations (7) and (8) demonstrate how magnetite can be formed from meteoric solutions.

Obviously magnetite will not form in the presence of free sulphuric acid. Further, sulphuric and carbonic acids are not generally assumed to remain as such in the presence of each other, especially when there exists an abundance of iron oxides and hydroxides, manganese oxide, silica, lime, magnesia, and colloidal substances such as are commonly found in erosional products.

It is believed that during the deposition of the magnetite in the Kinney Conglomerate a state of such delicate equilibrium existed that the ferrous and ferric salts were kept in solution until precipitation took place in the form of magnetite.

To substantiate this chemical hypothesis, a 0.25 molar solution of ferrous and ferric sulphate was mixed. A precipitate [18] was

[17] "According to S. Hilfert, the formation of the magnetic ferrosic oxide from non-magnetic hydrated ferrous and ferric oxides shows that the magnetic property lies primarily in the combination of the two—where ferric oxide is an acid anhydride and ferrous oxide is a base. Mellor, J. W.: A comprehensive treatise on inorganic and theoretical chemistry, 11: 732.

[18] Many experiments forming artificial magnetite under low temperature and

then formed by adding ammonium hydroxide. The precipitate was black and magnetic. The temperature was low, and the pressure was low; yet the product was magnetic.

FIG. 1.

An apparatus was set up as shown in Fig. 1. The ferric sulphate and ferrous chloride were allowed to seep drop by drop

pressure have been performed. J. W. Mellor (A comprehensive treatise on inorganic and theoretical chemistry. XIII, Fe, Part 2, p. 739) states that:
"R. Phillips found that if a mixture of ferrous sulphate and sodium carbonate be treated with sufficient potassium chlorate added all at one time, hydrated ferric oxide is formed, but if added in small portions at a time, the black ferrosoic oxide is formed and is not affected by subsequent addition of the chlorate.—In C. F. Wulffing's process, as described by F. J. R. Carulla, some ammonia is added to a ferrous liquor—say, waste liquor used for pickling iron—and air is blown into the mixture to oxidize the lower oxide. More ammonia is added to decompose completely the ferrous salt in solution, and the combination of the two oxides is brought about by means of heat and steam, or air pressure. The product is used as a magnetic oxide paint for protecting iron from corrosion. According to F. A. Abel and C. L. Bloxam, when ammonia is added to solution of a ferrous salt, a white precipitate is formed if air be excluded; in contact with air the precipitate becomes bluish-green, dark green, and finally black. In this state the precipitate is insoluble in water. If a mixture of ferrous and ferric salts be similarly treated, two distinct precipitates are formed—namely, the bluish-white ferrous hydroxide, and the brown ferric hydroxide. On standing for a short time, or on boiling, a reaction takes place between the two hydroxides, and one of the magnetic oxides results."

into the bottle containing a weak solution of ammonia. A black magnetic precipitate formed, which under the X-ray shows the diffraction pattern characteristic of authentic magnetite.

SUMMARY AND CONCLUSIONS.

Laboratory experiments show that magnetite can be formed under ordinary room pressure and temperature. X-rays reveal that laboratory-formed magnetite has a pattern identical with that of magnetite from the Kinney Mine, as well as with magnetite from other sources. Field observation shows that the Kinney magnetite occurs in an iron-ore conglomerate—a conglomerate which is a mechanical concentrate of the weathered underlying iron formation; it is porous in structure and is overlain by shale or glacial debris.

Since the magnetite crystals are found in abundance in this porous conglomerate but are lacking elsewhere, the writer concludes that:

1. At some places the slowly moving, brackish waters, saturated with iron sulphates and percolating through or escaping from the shale into the conglomerate, encountered local conditions of low pressure and precipitated their dissolved iron as marcasite.

2. At other places the stagnant waters were saturated with ferrous hydrocarbonates, which, in the conglomerate, were precipitated as goethite, hematite, or siderite.

3. In certain areas of this conglomerate the sulphate waters mingled with chalybeate waters, producing a mixture of ferrous and ferric salts in the proper proportions to form magnetite. The presence of phosphates, fossils, carbonaceous material, and manganese caused the reaction to be completed.

These conclusions are not contrary to accepted geologic beliefs.

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MICHIGAN COLLEGE OF MINING AND TECHNOLOGY,
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THE ORIGIN OF PRIMARY LEAD ORES: PAPER II.

ARTHUR HOLMES.

ABSTRACT.

In a former paper evidence was presented from which the writer inferred that ore-lead cannot be a derivative from either granitic or basaltic sources. To circumvent this heterodox conclusion Knopf suggests an alternative interpretation of the data, while Graton considers the data and underlying assumptions to be insufficiently well established to warrant so definite a conclusion. Keevil presents new data which he interprets to mean that the writer's conclusion is unnecessary. Knopf's suggestion that ore-lead is a differentiate from deep-seated peridotite, *via* basaltic and granitic magmas, is shown to be without adequate foundation. Much of Graton's criticism can be satisfactorily answered, but, in so far as some of the data are now known to be uncertain, it is frankly agreed that the conclusion must be regarded as one of probability, not certainty. On existing data, including the recent determinations by Keevil, for which high accuracy is claimed, the range of calculated atomic weights for different samples of granitic lead is significantly wider than the narrow range of determined atomic weights of ore-lead. Some samples of granitic lead have atomic weights that fall within the range for ore-lead, but it is pointed out that unless the corresponding granites are abnormally rich in lead there is no reason to suppose that they could actually yield deposits of ore-lead. The possibility that there may be abnormally lead-rich granites is, so far, unsupported by relevant data. Since it is improbable that ore-lead has been derived from granitic sources and, for similar reasons, still more improbable that it has come from basaltic sources, it becomes correspondingly probable that the source of primary lead ores lies within the substratum. In response to questions by Graton some speculative suggestions are offered bearing on the separation of ore-lead from the substratum, its migration to the upper levels of the crust, and the association of lead ores with granitic rocks rather than with peridotites.

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INTRODUCTION.

In my paper on "The Origin of Primary Lead Ores"[1] I calculated the average present-day atomic weights of granitic rock-lead and basaltic rock-lead, using data which I tacitly accepted as reliable for the purpose. The results revealed a highly significant contrast between the widely varying range of atomic weights calculated for granitic and basaltic rock-lead and the approximate constancy of the determined atomic weights of ore-lead. A similar contrast between the determined atomic weight of cotunnite-lead from Vesuvius and that of ore-lead had already been recorded and was cited as confirmatory evidence. I therefore concluded: "The proof is complete that ore-lead cannot be a concentration from either granitic or basaltic rocks, or from their respective magmas."

In his discussion of my paper Professor A. Knopf[2] finds no reason to criticize the data on which my demonstration was based, but he proposes an alternative petrogenetic interpretation of the evidence that would theoretically dispose of the problem, provided —as seems to be reasonable—that ore-lead and peridotitic rock-lead share the same limited range of atomic weight values. This alternative had seemed to me to be so untenable from a petrological point of view that I did not previously mention it as a practical possibility. Now, of course, I must pay it the respect of a systematic discussion.

[1] Holmes, A., ECON. GEOL., 32: 764–782, 1937.

[2] Knopf, A.: The origin of primary lead ores. ECON. GEOL., 32: 1061–1064, 1937.

In a searching and highly critical commentary on the topics of my paper Professor L. C. Graton[3] not only summarily rejects Knopf's attempt to solve the problem, but maintains that, for far more fundamental reasons, there may not even be a problem. With characteristic vigour he attempts to cut away the very roots of my conclusion by calling into question (*a*) the accuracy and representative character of the data I adopted;[4] and (*b*) the validity of the underlying assumptions implied in the method of calculation.

I am grateful to my friendly opponents for providing me with so many excellent reasons for examining more critically the subject matter of the discussion. It is clear that in one respect I understated the significance of the evidence on which I relied. Instead of being content, for the sake of brevity and logical directness, to work with and draw inferences from the average atomic weight of granitic (or basaltic) rock-lead, I should have developed the meaning of the *average*, with special reference to the range of values that any average necessarily implies. On the other hand, it is equally clear that I overstated the degree of certainty that could properly be ascribed to the conclusions I expressed. The probability that a generalization inferred from experimental data is true depends on the accuracy and representative character of the data and cannot be expected to attain the absolute certainty implied by the unqualified use of the term "proof." Instead of stating: "The proof is complete . . .", I should therefore have written: "On the data now available the probability is high that, in general, ore-lead cannot be a concentration from either granitic or basaltic rocks, or from their respective magmas." Since my paper was published—and partly

[3] Graton, L. C.: Ores: from magmas, or deeper ? A reply to Arthur Holmes. ECON. GEOL., 33: 251–286, 1938.

[4] Since submitting the present communication for publication, "the accuracy and representative character of the data I adopted" have been more specifically called in question by Dr. N. B. Keevil (Thorium-uranium ratios of rocks and their relation to lead ore genesis. ECON. GEOL., 33: 685–696, 1938). By the courteous permission of the Editor, to whom I am deeply grateful, I have been given an opportunity to incorporate a reply to Keevil's criticism. My comments on his results and conclusions will be found on pages 849–851.

because of its publication—some of the data on which I relied have been shown to be faulty or in need of confirmation. Consequently, the present probability of the above hypothesis no longer approaches the ideal of proof so closely as I had imagined.

Had I been without the apparent support of the experimentally determined atomic weight of the cotunnite-lead from Vesuvius, I should naturally have worded the conclusion with corresponding scientific caution. As it happens, my use of the cotunnite determination led Professor G. P. Baxter to recompute Piutti and Migliacci's results on the basis of present-day standards. He finds (personal communication) that for cotunnite-lead the atomic weight should be 207.67 instead of the recorded value 207.050; and that for common lead (determined as a check) it should be 207.81 instead of the recorded value 207.192. It may be noted that the original difference of .14 between the two atomic weights still remains. If this difference be genuine, as it well may be, it continues to support my inference that there should usually be a detectable difference between ore-lead and rock-lead. But the figures, as revised by Baxter, depart so widely from all other determinations that they are quite unacceptable. Thus, the difference between them has itself become open to suspicion. It is gratifying to know, however, that the publication of my paper, by the very challenge of its conclusions, has already quickened the development of plans for carrying out reliable determinations of the atomic weight and isotopic constitution of samples of rock-lead of known provenance.

Other doubts which have been expressed as to the reliability of the data involved in estimating the atomic weights of rock-lead will be discussed in the appropriate sections of the present communication.

EXAMINATION OF THE DIFFERENTIATION HYPOTHESIS PROPOSED BY KNOPF.

The Differentiation "Bridge."

The conclusion that the lead of lead ores has ascended from depths below the sialic and basaltic layers of the crust is ac-

cepted by Knopf, and so far we are in agreement. But I doubt the orthodox view that lead ores are derivatives from granitic (or related) magmas, and think it more likely that the lead travelled directly from the substratum as part of a procession of emanations, which, guided by deep-seated fractures, ascended to the upper part of the crust, where the ores were deposited. Knopf, on the other hand, seeks to reconcile a deep-seated source for lead ores with "the widely accepted idea of the genetic dependence of lead ores on granitic magmas." He holds that the lead travelled from the substratum as a constituent of magma that differentiated towards a granitic residuum, and that is was by further differentiation from this residuum that the lead was ultimately liberated as a constituent of ore-making emanations. He avoids the alleged atomic weight discrepancy by means of the following petrogenetic scheme:

> In terms of the problem here discussed, the ore-bringing magma of each metallogenic epoch was derived *de novo* from a basaltic magma, which itself was newly derived early in the petrogenic epoch from more basic material. This basic material was the result of selective fusion of deep-seated peridotite analogous in composition to meteoritic material. This source material is relatively rich in ordinary lead and is so poor in radioactive matter that despite the great age of the Earth, the minute amount of radiogenic lead formed and added to the lead primordially present has been insufficient to cause any appreciable deviation from the normal atomic weight 207.21 . . . During the course of each petrogenic epoch the lead, thorium and uranium, in accordance with the laws of magmatic differentiation, became concentrated in the granitic fraction, but the length of time of any of these petrogenic cycles was far too short for the lead radiogenically formed during any of them to have an appreciable effect on the atomic weight of the lead.

Graton dismisses this simple solution on the grounds that it is speculative and that the differentiation-mechanism implies the production of a relatively large volume of hypothetical material representing the pole complementary to the granitic end-product. It is doubtful whether either of these general criticisms will seriously weaken the faith of the thorough-going differentiationists. The differentiation solution is likely to prove very attractive, because of its economy in thought, unless its weaknesses are pointed out. Knopf's hypothesis is like a bridge of several spans connecting the dispersed lead of deep-seated peridotite with

the concentrated ores of the upper crust. Are all the spans adequately supported at each of their ends? If they are not, as I shall try to show, and even if a single span fails, then the bridge breaks down.

The Source-Material.

Knopf assumes that the source of lead is a deep-seated feldspathic pyroxene-peridotite or olivine-pyroxenite. There is no evidence that the substratum has such a composition. Analogy with stony meteorites, though generally regarded as not unreasonable, is strictly speaking, not evidence at all. The plain fact is that the composition of the substratum is not known. The position is the same as if only the refractive index of an otherwise unknown substance were known, and so no discrimination could be made between, let us say, cryolite and water. But in such a case it could at least be said that the substance was not fluorite. Seismological evidence indicates a possible identification of the material of the upper part of the substratum with dunite. But this possibility—though in itself not very helpful—constitutes evidence against an identification with peridotite of the type assumed by Knopf.

Passage from Peridòtite to Basaltic Magma.

Peridotites have been variously ascribed to (*a*) crystal accumulation from basaltic magma; (*b*) consolidation of peridotitic magma; and (*c*) metasomatic replacement of pre-existing rocks *e.g.* the hortonolite-dunite pipes of the Bushveld). Whereas (*a*) was formerly regarded as the dominant mode of origin, the greater importance of (*b*) is now being increasingly recognized. In an enormous work, of which four-fifths is devoted to the petrological problems associated with peridotites and serpentinites, Lodochnikov [5] records ample evidence for the independent existence of ultrabasic magmas (independent, that is, of basaltic

[5] Lodochnikov, V. N.: Serpentine und Serpentinite der Iltschirlagerstätte und im Allgemeinen und damit verbundene petrologische Probleme. Trans. Centr. Geol. Prosp. Inst., Leningrad, No. 38, 817 pp., 1936 (Russian with German Summary; see also Min. Abs., 7: 3–5, 1938.)

magma). He ascribes serpentinization to the concentration of the volatiles that characterize such magmas. From the associated veins (*e.g.* albitite, plumasite and rodingite) and contact metamorphic zones, he shows that the residual products of differentiation give rise to such minerals as grossularite, andradite, corundum, and albite. There is no suggestion that basaltic magma is a possible differentiation product. Hess [6] has recently summarized an extensive study of the ultrabasic intrusives that "occur abundantly in the present island arcs and in all the major orogenic belts of the world." He points out that large complexes of peridotite occur "without other igneous rocks of the same period of igneous activity." Referring to the differentiation of peridotite magmas, he writes: "Very small segregations of allivalite (bytownite plus olivine), grading into picrite and hence to peridotite, are found in the large intrusives, but have a minute size compared to the bulk of the intrusive." Neither of these investigators finds any evidence that basaltic magma is a differentiate from ultrabasic magma. Since selective fusion of peridotite would give the same result as fractional crystallization of peridotite magma,[7] there is clearly no natural evidence to support the hypothesis that basaltic magma has been derived from peridotite.

Passage from Basaltic Magma to Granitic Magma.

According to a widely held hypothesis, large bodies of granitic rocks are supposed to have consolidated from granitic magmas, which themselves were liquid differentiates from basaltic magma. I know of no evidence that can be said, even in its local application, to have raised this supposition from the intangible and unproved status of a theoretical possibility.[8] Occasional facts may

[6] Hess, H. H.: A primary ultramafic magma. Trans. Am. Geophys. Union, 18th. Ann. Meet., pp. 247–249, 1937. A primary peridotite magma. Am. Jour. Sci. (5), 35: 321–344, 1938.

[7] Bowen, N. L.: The Evolution of the Igneous Rocks, p. 316, 1928.

[8] Holmes, A.: The problem of the association of acid and basic rocks in central complexes. Geol. Mag., 68: 241–255, 1931. The origin of igneous rocks. Geol. Mag., 69: 543–558, 1932. The idea of contrasted differentiation. Geol. Mag., 73: 228–238, 1936; and 74: 189–191, 1937.

be consistent with it; the great bulk of the relevant evidence is not. In every country where petrologists are active, more and more occurrences of granitic rocks are every year being recognised as the results of such processes as metasomatism, rheomorphism,[9] and assimilation. It will suffice here to give a list of selected references.[10] It may be wrong, as Levinson-Lessing has

[9] Backlund has proposed the very useful term *rheomorphism* for the process whereby a pre-existing rock becomes partially or completely mobilized or fused as a result of the introduction of migrating materials (in great or small amounts) with concomitant rise of temperature.

[10] Anderson, A. L.: Contact phenomena associated with the Cassia Batholith, Idaho. Jour. Geol., 42: 376–392, 1934. Anderson, G. H.: Granitization, albitization, and related phenomena in the Northern Inyo Range of California-Nevada. Geol. Soc. Am. Bull., 48: 1–74, 1937. Backlund, H. G.: The problems of the Rapakivi granites. Jour. Geol., 46: 339–396, 1938. Barth, T. F. W.: Structural and petrologic studies in Dutchess County, New York: Part II: petrology and metamorphism of the Paleozoic rocks. Geol. Soc. Am. Bull., 47: 775–850, 1936. (Discussion, pp. 2005–2008.) Bemmelen, R. W. van: On the origin of the Pacific magma types in the volcanic inner arc of the Soenda Mountain System. De Ingenieur in Neder.-Indie (4), Mijnbouw en Geol., 4: 1–15, 1938. Collins, W. H.: Derivations of granitic rocks. Rep. 16th. Intern. Geol. Cong. (1933), 1: 271–282, 1936. Ebert, H.: Hornfelsbildung und Anatexis im Lausitzer Massiv. Z. d. Deutsch. Geol. Ges., 87: 129, 1935. Fenner, C. N.: Some magmatic problems. Jour. Wash. Acad. Sci., 24: 113–124, 1934. A view of magmatic differentiation. Jour. Geol., 45: 158–168, 1937. Goodspeed, G. E.: Small granodiorite blocks formed by additive metamorphism. Jour. Geol., 45: 741–762, 1937. Goodspeed, G. E. and Coombs, H. A.: Replacement breccias of the Lower Keechelus. Am. Jour. Sci. (5), 34: 12–23, 1937. Holmes, A.: Transfusion of quartz xenoliths in alkali basic and ultrabasic lavas, Southwest Uganda. Min. Mag., 24: 408–421, 1936. Holmes, A., and Harwood, H. F.: The volcanic area of Bufumbira. Geol. Surv. Uganda, Mem. 3, Part II: 243–285, 1936–1937. Horwood, H. C.: Granitization in the Cross Lake Region, Manitoba. Trans. Roy. Soc. Can. 3rd. ser. (IV), 30: 99–117, 1936. Larsen, E. S. and Morris, F. K.: Origin of the schists and granite of the Wachusett-Coldbrook Tunnel, Massachusetts. Geol. Soc. Am. Abst., Dec. 1928, p. 26. MacGregor, M.: The Western Part of the Criffell-Dalbeattie igneous complex. Quart. Jour. Geol. Soc., London, 93: 457–486, 1937. Quirk, T. T. and Collins, W. H.: The disappearance of the Huronian. Geol. Surv. Canada, Mem. 160, 1930. Read, H. H.: The petrology of the Arnage district in Aberdeenshire: A study of assimilation. Quart. Jour. Geol. Soc., London, 79: 446–486, 1923. Reinhard, M.: Gesteinsmetamorphose in den Alpen. Jaarb. v. d. Mijnb. Ver. Delft, 1934–1935, pp. 39–45. Reynolds, Doris, L.: The eastern end of the Newry Complex. Quart. Journ. Geol. Soc., London, 90: 585–636, 1934. The Two Monzonitic series of the Newry Complex. Geol. Mag., 73: 337–364, 1936. Demonstrations in petrogenesis from Kiloran Bay, Colonsay. I. The transfusion of quartzite. Min. Mag., 24: 367–407, 1936. Contact phenomena indicating a tertiary age for the gabbros of the Slieve Gullion Dis-

warned us, to generalize from such evidence, but if so, it is surely much more seriously wrong to generalize from no evidence at all and claim that granites are differentiates from basaltic magma.

It may also be noted that the differentiation bridge from peridotite to granite fails to provide the necessary amount of granitic material for the continental crust. On the estimate most favorable to granitic material, the hypothesis provides basaltic and granitic materials in the proportion 10:1, whereas what is needed is about 10:6. Beneath considerable areas of the oceanic floor, however, little or no granitic material appears to have been generated, despite the apparent availability of ample supplies of basaltic magmas. These discrepancies are too serious to be lightly swept aside. The fact remains that, so far back as geological history has penetrated, granitic rocks appear in bulk *only* where pre-existing sialic rocks (including siliceous and argillaceous sediments) already existed, or (as in the case of Iceland) can be inferred to be present under a basaltic capping. It serves no useful purpose to enquire where the first granites came from, since at present we have no idea what the composition of the original crust may have been.

Among many geochemical difficulties that the differentiation bridge fails to resolve is one with a special bearing on the subject matter of this paper. Ultrabasic rocks appear to be relatively rich in lead, as Knopf points out, whereas, of all the common rock groups those of basaltic composition are the poorest in lead. If the passage of potential granitic material from peridotite into basaltic magma is accompanied by a marked impoverishment in lead, it is difficult to believe that a marked concentration of lead could take place in the next stage, from basaltic magma to granitic magma.

trict. Proc. Geol. Assoc., London, 48: 247–275, 1937. Sederholm, J. J.: On migmatites and associated Pre-Cambrian rocks of Southwestern Finland. Bull. Comm. géol. Finlande, 58, 1923; 77, 1926; and 91, 1930. Stark, J. T.: Migmatites of the Sawatch Range, Colorado. Jour. Geol., 43: 1–26, 1935. Wegmann, E.: Zur Deutung der Migmatite. Geol. Rundsch., 26: 305–350, 1935. Geological investigations in Southern Greenland: Part I. On the strctural divisions of Southern Greenland. Medd. om Grønland, 113: No. 2, 148 pp., 1938.

Passage from Granitic Magma to Lead Ores.

It is widely assumed that the common association of lead ores with granitic rocks implies a genetic relationship of the daughter-mother type. This assumption seems to be unsupported by any evidence independent of the association itself. The probability of its validity is gravely reduced by the fact that this association is far from being systematic, and by other observed facts relating to the relationship of lead ores to granitic rocks in space and time.

If the relationship were of the daughter-mother type, then—where lead ores occur near granitic centres of the same cycle—the material of the lead ores and of the associated ores should have been concentrated in the residual fractions of the former granitic magmas. In such areas pegmatites should, therefore, pass laterally (along the strike) or vertically (up the dip) into mineral veins containing lead ores. Some pegmatites contain tin ores, and the passage of pegmatites into quartz veins has occasionally been recorded. Lindgren, however, questions "whether anybody has ever seen a pegmatite dike running over in continuation into a tin vein . . . or a pegmatite dike running over into an economically important gold quartz vein."[11] If the evidence for tin and gold is so doubtful, that for lead and its associates is practically non-existent. As a constituent of pegmatites and of the veins into which they pass, galena is only a mineralogical curiosity. Lead veins of economic importance are of later formation than the pegmatites of the same cycle. Their constituents were not *in* the pegmatite "magma"; they followed up from behind at a later stage. If these constituents share a granitic parentage with pegmatites, one may well ask how they came to be so effectively left behind in the interstices of the deeper parts of a granitic mass at the time when the pegmatitic material migrated outwards. Unless this question—and the far wider general question raised by ore sequences in general—can be satisfactorily met, it can only be said that lead ores come from an unknown source in the depths. To correlate that source with the

[11] Lindgren, W.: Magmas, dikes and veins. A. I. M. E., Pam. 1575–I, p. 27. 1936.

deeper parts of a granitic mass is to favor one possibility out of several, while ignoring the special difficulties that it fails to meet.

Necessity for Further Evidence.

The foregoing survey of Knopf's bridge hypothesis has revealed lack of support at every point where support is necessary, if the bridge is to stand. Graton's summary dismissal of the hypothesis is completely justified.

It will be gathered, however, that for purely geological reasons I question the validity of Graton's specific inferences from what he calls (p. 275) the "significant geological connections between ores and magmas." I fully realize, of course, that it would be wrong to infer that primary lead ores cannot be derivatives from granitic material merely because I fail to understand in the light of present knowledge *how* they could have been so derived. To answer the question whether the emanations from which primary lead ores are deposited are liberated from some source other than granitic obviously requires further evidence, such as may be provided by determinations of the atomic weight and isotopic constitution of lead.

It may be noted that atomic weight evidence cannot prove that lead ores are derivatives from granitic material; it may, however, provide evidence indicating that they are not so derived in specific cases and the probability that they are not in general. A year ago I thought the data sufficient to raise that probability to practical certainty. The present doubts as to the reliability of the data, to which Graton has very properly directed attention, indicate the need, not only for more accurate data, but also for greater caution in interpretation.

EXAMINATION OF DATA AND ASSUMPTIONS.

Development of the Atomic Weight Argument.

The greater part of Graton's paper is an attempt to show that my atomic weight argument against a granitic source for lead ores is not conclusive. Much of his criticism is dependent on

his assumption that 207.14 is the calculated value of the present-day atomic weight of granitic rock-lead. Actually, this figure represents the calculated average atomic weight and is therefore only one particular example of a range of values. Reference to Fig. 1 of my 1937 paper (p. 773) shows not only that Ra and Th vary in concentration, but also that the ratio Th/Ra or Th/U varies between wide limits in granitic rocks. Fig. 2 (p. 775) illustrates similar variations for basaltic rocks. Since the ratio Th/U controls the proportion of thorium-lead to uranium-lead generated in rock material, it follows that the atomic weight of the radiogenic lead formed in different samples must also vary between corresponding limits. The total amount of lead (*i.e.* of rock-lead) in each of these samples is unknown, but no serious error is likely to be introduced by assuming that each sample contains the average amount proper to granitic rocks (or basaltic rocks) as a whole. On this assumption the figures in Table I

TABLE I.

CALCULATED ATOMIC WEIGHTS OF ROCK-LEAD.

Present Value of Th/U	Granitic Rocks Pb = 30 × 10^{-6} gm./gm.		Basaltic Rocks Pb = 5 × 10^{-6} gm./gm.	
	U = 6 × 10^{-6} gm./gm.	U = 4 × 10^{-6} gm./gm.	U = 1.8 × 10^{-6} gm./gm.	U = 1 × 10^{-6} gm./gm.
0	207.11	207.14	207.02	207.11
1	207.125	207.15	207.06	207.125
1.7	**207.14**	207.16	—	—
2	207.145	207.17	207.09	207.145
2.4	—	—	**207.10**	207.15
3	207.16	207.18	207.13	207.16
4	207.18	207.19	207.16	207.18
5	207.20	207.20	207.19	207.20
5.6	207.21	207.21	207.21	207.21
6	207.22	207.215	207.22	207.22
7	207.24	207.23	207.26	207.24

have been calculated to illustrate the present-day range of atomic weight values for rock-lead in samples of granitic and basaltic rocks containing different proportions of U and Th.* The figures in heavy type are the averages used in my 1937 paper.

* I am indebted to my friend Dr. W. D. Urry for bringing to my attention a very elegant graphical method which he has devised with a view to facilitating such cal-

It will be noticed that when the value of Th/U lies between 5 and 6 the calculated atomic weight of the resulting rock-lead ($207.21 \pm .01$) is indistinguishable from the atomic weight of ore-lead. When the value of Th/U is less than 5 the calculated atomic weight of the rock-lead is less than the atomic weight of ore-lead, and for any given value of Th/U the atomic weight

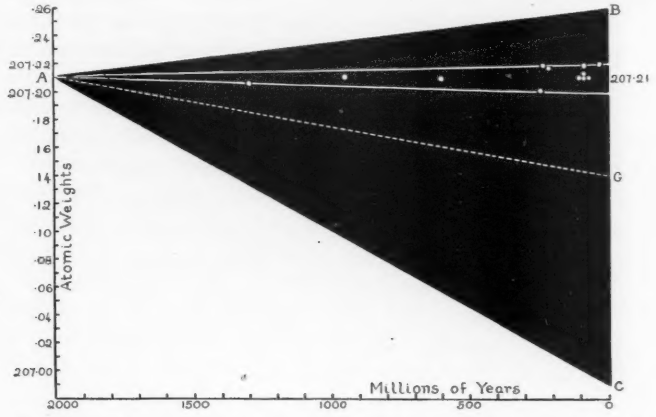

FIG. 1. Diagram illustrating the contrast between the atomic weights of various samples of ore-lead of known geological age (white dots in wedge) and the range of calculated atomic weights of granitic rock-lead ("fan," *ABC*). Data as in Fig. 1 of 1937 paper. The average calculated atomic weight of granitic rock-lead is represented by *G*, and its variation during geological time by the dotted line *AG*.

deficiency increases with the proportion of U present in the rock. Similarly, for values of Th/U greater than 6 the calculated atomic weight of the rock-lead is greater than the atomic weight of ore-lead.

Taking the actual data plotted in Fig. 1 (1937, p. 773), the

culations. In applying this method, however, I have avoided the uncertainties in the quantitative relationships between uranium I and actino-uranium by adopting the value 206.00 for the atomic weight of uranium-lead. The determined values for the atomic weight of uranium-lead range from 205.93 to 206.01 (see 1937 paper, p. 771).

calculated atomic weight values for the granitic rock-lead of the individual samples are found to range between 207.26 and 206.99. The weighted average of these extreme results and those falling between is 207.14. A graphic representation of the present-day range is reproduced in the adjoining diagram, Fig. 1. The calculated present-day range appears vertically on the right (*BC*). For successively earlier geological periods the range gradually narrows until, at the beginning of geological time—at the left of the diagram—it reduces to the point *A*, representing the assumed atomic weight of the earth's primeval lead, here taken at 207.21. All the possible values for the atomic weight of granitic rock-lead during geological time—on the data here adopted—fall into the "fan" shown in the diagram by the black triangle, *ABC*.

If ore-lead represents a concentration of granitic rock-lead it would be expected that the atomic weights of samples of ore-lead of different geological ages and from different sources should be scattered through the fan, with crowding about the "average" line *AG* and especially about the right-hand quarter of the line (since most commercial lead is from ores of late Pre-Cambrian or later age). The determined atomic weights of samples of ore-lead of known geological age are indicated in Fig. 1 by white spots. Like the more numerous determined atomic weights of ore-lead of unknown provenance, they never depart far from 207.21. The white spots themselves fall within a very narrow fan or wedge-like area, delimited by white lines. The scatter to be expected on the hypothesis that ore-lead is a concentrated extract of granitic lead is obviously not realized.

For basaltic rocks (Fig. 2, 1937) a similar fan with a similar "spread" can be constructed. Again there is a marked contrast between the broad rock-lead fan and the narrow ore-lead wedge.

In both cases, however, the rock-lead fan encloses the ore-lead wedge. It therefore follows that a few rocks—representing a very small percentage of the whole—contain lead having an atomic weight indistinguishable from that of ore-lead. At this point the demonstration obviously falls short of complete "proof," and to this extent Graton's criticism is thoroughly justi-

fied. It may be pointed out, however, that except in the case of extremely lead-rich magmas (see p. 852) there is no reason to suppose that the processes of differentiation and concentration alleged to be responsible for the formation of lead ores from granitic magma operated only in the few cases where the atomic weight of the lead happened to be $207.21 \pm .01$. So far as the latter figure is a consequence of the value of Th/U it cannot possibly have had any bearing on the course of differentiation. If ore-lead cannot have come from most granites, it thus becomes highly improbable that it can have come from any, unless certain granites are especially favoured by containing an abnormally high proportion of lead.

It is now necessary to examine the data used, and the various assumptions made in the course of the above argument.

Lead Data.

On p. 260 of his paper Graton objects to my use of the average content of lead in granitic rocks (30×10^{-6} gm./gm., as determined by Hevesy and Hobbie), on the ground that, as the sampling is likely to be imperfect, the average "cannot be held reasonably to typify all granitic rocks, but only such specimens in such proportion of varieties as were available to the analysts' hand." I should readily accept the objection if there were any evidence (*a*) that the sampling was imperfect, or (*b*) that the range of the lead content of granitic rocks was sufficiently wide to invalidate the argument.

The rocks in the composite sample analyzed by Hevesy and Hobbie comprise granites of various geological ages from and including the Pre-Cambrian to the Tertiary, representing all the chief orogenic cycles of Europe and North America and including a few from Africa and Asia. As a petrologist, I am personally satisfied that the sampling was sufficiently representative both for their purpose and mine. It would, of course, be desirable to have similar data for every continent, but there is no geochemical reason to suspect that the granites of Africa or Australia, for example, should contain appreciably more lead than those of Europe or North America. The geochemical variations

in space and time within a single continent are generally found to be characteristic of all.

It is readily seen that if some granitic rocks contain many times as much lead as the Hevesy and Hobbie average—up to twenty times as much being necessary—then, in those rocks, the proportion of radiogenic lead would be relatively too low to make any significant difference to the resultant atomic weight of the total rock-lead. The fan of Fig. 1 might then effectually contract towards the ore-lead wedge. On the other hand, with a relatively low content of total lead the effect of radiogenic lead becomes proportionately greater and the spread of the fan is correspondingly increased. Hence the importance of knowing the range which may be hidden in the average.

The data for all the individual samples of granitic rocks that have been analyzed for lead are listed in Table II. Many of the results refer to rocks that have been regarded as representatives of the magmas from which lead ores of economic importance have been derived. It will be seen at once that, despite this fact, the actual lead content never departs far from the average determined by Hevesy and Hobbie. Moreover, the maximum values recorded are probably too high to be "reasonably" representative. Describing the pyritiferous porphyry of Leadville, Emmons[12] writes: "crystals of pyrite are frequently concentrated along cleavage planes [i.e. of biotite or chlorite] sometimes associated with finely disseminated crystals of galena." Such introduced lead should not, of course, be regarded as part of the rock-lead proper to the rock just before the time of ore deposition. Indeed, Graton himself, referring to the 58 samples of granitic rocks that Hevesy and Hobbie analyzed, points out (p. 270):

> If a single one of these samples had contained in each gram enough tiny scattered particles to aggregate the equivalent of a cube of galena less than 0.3 mm. on a side, this one sample would account for the whole of the 30 parts per million of lead found in the entire 58 specimens.

It thus appears that if there is any systematic error (from the point of view of the present discussion) involved in the average

[12] Emmons, S. F.: Geology and mining industry of Leadville, Colorado. U. S. G. S. Mon. 12: 82, 1886. On p. 327, W. Cross writes: "Galena appears locally in small quantity, but only on fissure planes."

adopted, it is most likely to be in the direction of making that average too high to be typical of granitic rocks as a whole. The results compiled in Table II, however, suggest that the average is representative and that the range is small. Even including the suspect Leadville rocks, the Table II average is 34 parts per million, in good agreement with Hevesy and Hobbie's 30 parts per million. Without the Leadville rocks, but still including several from provinces of lead ores of the same magmatic cycle, the average is only 27. Even if the average were as high as 60 parts per million, the fan of Fig. 1 would still have a present-day spread from 207.10 to 207.24, with 207.175 as its weighted average.

TABLE II.

THE LEAD CONTENT OF GRANITIC ROCKS.

Rock and Locality	Lead gm./ 10^6 gm.	Analyst and Reference
Leadville, Colorado.[13]		
Gray Quartz-porphyry, Onata Mine	54	Hillebrand, W. F.: U. S. G. S. Mon. 12: 591, 1886.
Pyritiferous Quartz-porphyry, White's Hill	39	
Do., Head of White's Gulch	60	
Pre-Cambrian, Missouri.		
Granite, Graniteville, St. Francois Co.	24	Robertson, J. D.: Missouri Geol. Surv., 7: 479, 1894.
Granite, Clearwater, Reynolds Co.	20	
British Isles.		
Granite, Foxdale, Isle of Man	40	Finlayson, A. M.: Q. J. G. S., 66: 301, 1910.
Granite, Threlkeld, Lake District	30	
Granite, Glendalough, Co. Wicklow, Eire	40	
Granite, Dartmoor, Devonshire	20	
Alaska.		
Rhyolite Pumice, Katmai, 1912	19	Zies, E. G.: Nat. Geog. Soc. Tech. Pap., Katmai Series, I (4), p. 58, 1929.
Rhyolite Glass, Novarupta	26	
Composite of 58 samples.	30	Hevesy, G. and Hobbie, R.: Nature, 128: 1038, 1931.

[13] It should be noted that 13 of Hillebrand's 16 results for acid rocks refer only to lead present as sulphide. In the three here cited, the "insoluble" lead was also determined. The average of the 16 results, recorded as PbO, is Pb = 20 gm. per 10^6 gm. of rock.

It is of interest to notice that to reduce the maximum atomic weight of 207.26 (*B* in Fig. 1) to 207.22 the rock concerned, if

otherwise of average type, would have to contain 148 parts per million of ordinary lead. This figure is very much higher than any yet found, even including the probably contaminated Leadville rocks. At the point *C* of Fig. 1 the minimum atomic weight is 206.99. To increase this to 207.20 the content of ordinary lead would have to be 640 parts per million. Now if high figures be admitted for some of the individual specimens, then obviously most of the others must have a lead content far below the average, in which case the fan not only remains, but may even be considerably increased in spread.

Graton suggests that "there appears to be a distinct tendency for Pb, Th and U in rocks to vary in the same direction" (p. 259). If this were so, then high radiogenic lead might be effectually diluted by high ordinary lead. But the geochemical data, as far as they go, lend no support to this suggestion. Ultrabasic rocks contain on an average less U and Th than basic rocks, but more Pb. Alkali granites and syenites contain on an average more U and Th than ordinary granites, but less Pb. Pegmatites characterized by thorium minerals tend to be poor in uranium minerals. Pitchblende deposits are poor in thorium. Iron meteorites are not rocks in the ordinary sense, but the fact that they are relatively rich in lead and exceptionally poor in U and Th is one of relevant significance.

It may be concluded that, unless the data for lead are wildly and hopelessly wrong—an assumption for which there is no support—my adoption of the average of Hevesy and Hobbie cannot be held to invalidate the argument. It may be urged, of course, that lead-rich granites may exist somewhere, though remaining undiscovered; that with these granites, or some of them, lead ores may be found in association; and that, in consequence, there may be room for the hypothesis that some lead ores may be derivatives from granitic magmas, even though most are not. To prove this hypothesis would be practically impossible with present resources, but to disprove it with absolute finality is obviously equally impossible. The impasse arises solely from a hypothetical possibility that may never materialize; while, if it did,

it could not be proved that it was not a case of coincidence, such as must occasionally happen.

A further point that may be conveniently disposed of here arises from Graton's assumption (p. 253) that my conclusion depends on the proposition: "That all the lead existing at the earth-beginning (*i.e.* primeval lead) was of uniform character and possessed the same atomic weight, 207.21, as now found for 'ore-lead' of all ages."

On p. 262 he states further that this premise "seems to be not soundly established, but only one presumption among many that could be made." Reference to Fig. 1 shows at once that it makes no difference to the argument whether the fan opens out from the point $A = 207.21$, or from some other higher or lower point. The fan, only slightly modified in area, merely swings to a higher position when A is $207.21 + x$ and to a lower position when A is $207.21 - y$. If A is variable, then the fan spreads from the line between $207.21 + x$ and $207.21 - y$, and is thus extended in area. Similarly if the duration of geological time is greater or less than 2,000 million years, the fan correspondingly expands or contracts a little. In any case a fan remains, and with it the contrast between the approximately constant atomic weight of ore-lead and the wide variability of granitic rock-lead. The conclusion clearly does not depend on the proposition that the earth's primeval lead had the same atomic weight as the ore-lead of today. The evidence suggests that it had, but this is not a necessary premise and therefore the premise need not be "soundly established." The fan remains.

The above section has been devoted specifically to granitic rock-lead, since the discussion centres on granitic lead rather than on basaltic lead. Similar considerations, however, apply with equal force to basaltic lead and need not be repeated.

Radioactive Data.

Graton (p. 257) states that "the data to be used in geological application of radioactivity are still in a state of flux and change"; and although he acknowledges "that the various kinds

and magnitudes of error . . . do not necessarily cause proportionate cumulative inaccuracy in the final result" (p. 258), he feels it necessary "to conclude that the atomic weight of rock lead cannot be reached with the precision necessary to validate the use Holmes now tries to make of it." When it is realized that the average atomic weight of rock-lead implies a broad fan of widely varying values (Fig. 1), it will be appreciated that no great precision was either claimed for that particular average or thought to be necessary.

Nevertheless Graton is perfectly right to draw attention to the disturbing possibility that some of the radioactive determinations may be considerably in error. Since my 1937 paper was written, it has become known that a systematic checking of standards and data is in progress at the Massachusetts Institute of Technology, and that certain remarkable discrepancies have already been disclosed. It appears that, possibly as a result of faulty standards, some of the published determinations of U in basaltic rocks are too low. Of more immediately serious importance is the discovery that determinations of U in granitic rocks made by counting alpha-particles are often lower than the older determinations made with the electroscope. No serious doubt appears to have been thrown on the corresponding Th determinations.

So far as basaltic rocks are concerned, there is clearly no need to discuss the changes further, since they strengthen rather than weaken the force of the original argument. For granitic rocks, however, the suggested changes are in the opposite direction. The M. I. T. results are not published at the moment of writing, but a recent paper by Poole [14] indicates the order of the difference involved. Granite from Mt. Manitou, Colorado, was found to have a Ra content of 2.1×10^{-12} gm./gm. by the electroscope method and 1.64×10^{-12} gm./gm. by the new method, using a direct fusion furnace and counting the alpha-particles. In terms of uranium the respective figures are 6.3×10^{-6} gm./gm. and 4.9×10^{-6} gm./gm. For other sialic rocks the differences are

[14] Poole, J. H. J.: A method for determining the radium content of rocks by direct alpha-ray counting. Sci. Proc. Roy. Dublin Soc., 21 (N. S.), No. 54: 595–608, 1938.

less than in this case. Poole notes that the cause of the discrepancies is unknown and requires further investigation. It may be noted that two other rocks, a basalt and an eclogite, gave the same results by both methods. It is, therefore, by no means established that the new results for granitic rocks are necessarily more correct than some, at least, of the older ones.

For the sake of the present discussion, however, it may be assumed that the U results for granitic rocks should be reduced throughout, so that the average content becomes 4 gm./10^6 gm. instead of 6 (see the third column of Table I). If the Th results remain as before, then the average value of Th/U becomes 2.5 instead of 1.7, and the calculated average atomic weight of granitic rock-lead becomes 207.173 instead of 207.14. There still remains a conspicuous atomic weight fan for granitic lead with a present-day spread from 207.07 to 207.25 (Fig. 2).

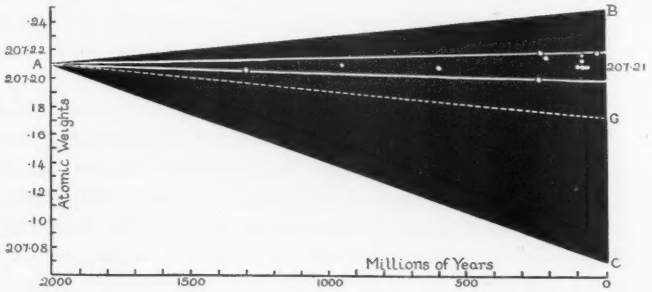

FIG. 2. Diagram illustrating the contrast between the atomic weight "wedge" for ore-lead and the calculated atomic weight "fan" for granitic rock-lead (*ABC*). Data as in Fig. 1, but with the uranium contents of the analyzed samples of granitic rocks reduced in the ratio 6:4. With the data so modified the average calculated atomic weight of granitic rock-lead is represented by *G*, and its variation in geological time by the dotted line *AG*.

Since writing the preceding paragraph, a further series of new results has been published,[15] from which it appears that the

[15] Keevil, N. B.: Thorium-Uranium ratios of rocks and their relation to lead ore genesis. ECON. GEOL., 33: 685–696, 1938.

reduction assumed in that paragraph may still be insufficient. From determinations on twelve specimens of North American granitic rocks Keevil finds the following average values:

$$\left.\begin{array}{l} U = 2.77 \text{ gm./}10^6 \text{ gm.} \\ Th = 7.94 \text{ gm./}10^6 \text{ gm.} \end{array}\right\} \frac{Th}{U} = 2.8$$

From these average data he calculates the present-day atomic weight of granitic lead to be 207.19. Assuming that the atomic weight of the earth's primeval lead was near 207.21, he gives the expected range in the atomic weight of granitic lead as from 207.21 in early geological time to 207.19 at present. The actual atomic weights of samples of ore-lead of known geological age (Figs. 1 and 2, and Keevil's Table IV) vary, though not systematically, from 207.205 in early geological time to 207.22 in Miocene time. Obviously, there is still a discrepancy, both numerically and in the direction of variation in time. Nevertheless, Keevil concludes that this discrepancy is so small that it "makes unnecessary the conclusion reached by Holmes that ore leads must come from a deep magmatic source."

The real discrepancy, however, is very much greater than appears from Keevil's calculations. Like Graton, Keevil has overlooked the atomic weight significance of the range of variation of the individual values of U, Th, and Th/U. The twelve sets of results for granitic rocks range between the following extremes, corresponding to which the expected atomic weights of the resulting granitic leads have been added:

Granitic Rock	West of Taschereau, Quebec. Pre-Cambrian	Chelmsford, Mass. Late Palaeozoic	Fitchburg, Mass. Late Palaeozoic
U	.58 gm./10^6 gm.	8.0	2.5
Th/U	1.5	1.7	4.2
Atomic weight of granitic lead	207.20	207.13	207.21

Thus, on Keevil's own determinations the expected variation in the atomic weight of granitic lead is not merely from 207.21 originally to 207.19 now, but is actually from 207.21 originally

to a present-day range of 207.13–207.21, which is a significantly different result. For basaltic rocks (including two monchiquites from Colonsay) the new results published by Keevil show that the expected present-day range for the atomic weights of basaltic lead is even greater than for granitic lead. Keevil's inferences (*a*) that my conclusion is unnecessary, and (*b*) that lead ores "are probably derived from basaltic or granitic rocks or their magmas," are thus found to be unsupported by his own evidence.

Keevil claims "that the differences expected by Holmes between the atomic weights of rock leads and ore leads are not verified." Quantitatively this is true, but for our immediate purpose it is of the utmost importance that it should be clearly realized that differences *do* still remain and that they are differences that should be easily detectable. The fact that they are less than I originally expected does not invalidate my argument.

The new results, like the partly discredited older ones, provide no reason for supposing that the atomic weight fan for granitic lead coincides with the narrow wedge (Fig. 2) which encloses the ore-lead atomic weights. The restricted variability of the latter (207.20–207.22) continues to remain unexplained, if ore-lead be assumed to have come from granitic magmas, unless it be further assumed that lead ores have been formed, not from granitic materials of any kind, but *only* from granitic material containing lead with an atomic weight near 207.21.

To be justified, this last assumption requires that one or other of three conditions should hold good for the supposed ore-generating granites:

(1) The time-average of the varying values of Th/U happened to be near 4 or 5 at the time of ore formation. The present value of Th/U for this condition is near 5 or 6. As already pointed out, it is difficult to believe that any process of differentiation that might be responsible for the separation of lead ores from granitic material could depend on the numerical relations between U and Th.

(2) The contents of U and Th are so low that the resulting radiogenic lead is negligible compared with the average content

of total lead. This condition again imposes a fantastic limitation on processes of differentiation.

(3) The content of total lead is exceptionally high compared with the average, without correspondingly high U and Th. This condition has been discussed on p. 846. It need only be repeated that all the existing data (Table II) indicate that the analyzed granitic rocks with which lead ores are associated have a lead content that is not exceptionally high, but of the same order as the average amount.

It must be noticed, however, that the excess of ordinary lead necessary to bring the atomic weight of the total lead within the limits for ore-lead decreases as Th/U approaches 5 or 6 (from either side), and decreases as the U and Th contents decrease. Thus, if the revised radioactive data for granitic rocks turn out to be correct, the proportion of granites containing lead of atomic weight appropriate to ore-lead and in approximately average amount will be greater than on the older data.

It does not follow, of course, that because such granites contain lead of the "right" atomic weight they actually did yield primary lead ores. There are three possibilities. Let us distinguish granites with lead of the right atomic weight as "X granites," referring to the others as "the remaining granites."

(*a*) Neither the X granites nor the remaining granites yielded lead ores. In this case the ores must have come from some source other than granitic.

(*b*) The X granites yielded some occurrences of primary lead ores, while other occurrences came from some source other than the remaining granites.

(*c*) The X granites yielded all occurrences of primary lead ores.

Possibilities (*b*) and (*c*) imply that the X granites could yield lead ores, but that the remaining granites could not. It cannot be supposed, however, that the atomic weight of a minute quantity of dispersed lead could in any way affect the course of differentiation, and consequently the remaining granites, if they exist, should be just as likely to have yielded lead ores as the X

granites. Since all the available evidence consistently indicates that the remaining granites have not yielded lead ores, it follows with a high degree of probability either (*a*) that the X granites have also failed to yield lead ores, or (*b*) that there are no remaining granites, *i.e.* that all granites are of the X type. Inference (*b*), however, is contradicted by the observed variations in the value of Th/U.

From the above analysis of the existing data it may be inferred that lead ores have probably not been derived from granitic materials. If it should turn out, beyond all expectation, that the existing data are hopelessly misleading and if further investigation shows that practically all granites belong to one or more of the three groups: (1) with Th/U near 5 or 6; (2) with very low U and Th; (3) with high Pb; then the above inference could no longer be drawn. The inference would not, on that account alone, be necessarily wrong, but if made, it would have to be drawn from other evidence.

Selective Extraction of Isotopes.

Graton makes the apparently reasonable suggestion (p. 266) that "the chance for selective diffusion of isotopes . . . must be greater in nature's laboratory than in man's." He goes on to ask whether there may not have been a selective extraction and concentration of the isotopes of rock-lead that would yield galena-lead with an average atomic weight of 207.21. Here, Graton appears to have overlooked the fact, experimentally established, that the lighter isotopes of a given element have a higher diffusion-coefficient than their heavier companions. Thus, the effect of diffusion on a procession of migrating lead isotopes would be to increase the proportion of the 206 and lighter isotopes in the extract, so that any lead deposited from the latter would have a lower atomic weight than the lead of the source-material. The Table on p. 265 of Graton's paper shows that he envisages the possibility of an ore-lead extract with 29 per cent of the 206 isotope forming from an average granitic rock-lead with 32.7 per

cent of the 206 isotope. Actually, if diffusion promoted isotopic fractionation the change would be in the opposite direction.

An objection to Graton's suggestion, which is of greater petrogenetic interest, arises from the astonishing discovery of recent years that in nature's laboratory selective diffusion of isotopes appears to be a negligible process. In his book on *Mass Spectra and Isotopes* (pp. 186–188), Aston presents a formidable array of evidence from which he infers "that the evolution of the elements must have been such as to lead to a proportionality of isotopes of the same element which was constant from the start." In the case of lead, this conclusion cannot be rigidly tested, because of the production of lead isotopes by radioactive disintegration during geological time. But in the case of many other elements, including N, Cl, Fe, Cu, Hg, Sb, and U,[16] no measurable variations in atomic weight or isotopic proportions have been detected throughout a wide range of terrestrial and (for some of the elements) meteoritic sources. Samples of chlorine (76 per cent of isotope 35 and 24 per cent of isotope 37) from many specimens of sea-salt, potash deposits, apatite, sodalite, scapolite, and meteorites have been very carefully investigated, but no differences have been detected. That there should be this constancy is amazing, since even in the laboratory selective concentration of the lighter isotope (35) of chlorine has been effected by diffusion, and the atomic weight thereby reduced from 35.456 to 35.418. If natural processes fail to bring about any detectable change in the atomic weight of chlorine, they are hardly likely to do so in that of lead. For chlorine the tendency of the isotopes to separate as a result of their different rates of diffusion involves the 36th root, whereas for lead the 207th root is involved. Thus Graton's appeal to selective extraction of isotopes can be dismissed not only because, if such fractionation did occur, it would generally act in the wrong direction for his requirements, but also because the process does not appear to be a natural one at all.

It might appear that atomic transport by diffusion cannot be a dominant process in crystal growth and replacement phenomena.

[16] Evans, R. D.: The determination of the age of iron and stony meteorites from their radioactivity. Pop. Astron., 46: 8, 1938.

Apparently, however, there may be a way out. Korzhinsky[17] has attempted to show that in the diffusion concerned in metasomatism the mobilities of the ions depend on the ionic radii of the migratory elements. It is probable that valency is also a factor. Thus, natural diffusion may not depend so much on the isotopic masses as on their volumes and valencies. Both of these are the same for the constituent isotopes of any given element and consequently detectable selective diffusion of isotopes—as distinct from elements—may not take place.

A brief digression may here be made in reference to a suggestion by Wells.[18] He attempts to explain away the atomic weight discrepancies "by assuming that radiogenic lead remains fixed in the uranium-thorium minerals and is not extracted by migrating waters to form ore deposits." The objection to this proposal is that both ordinary and radiogenic lead are dispersed through rock-forming minerals of all kinds. Extraction of lead from the latter by meteoric or hydrothermal solution could therefore not be selective. Hillebrand's analyses of the Leadville rocks show that only part of the lead in these rocks is present as sulphide. The latter would certainly be more soluble than the silicate minerals containing non-sulphide lead, but this consideration is of no help to Wells. If the sulphide-lead was not introduced, then it is granitic lead and the atomic weight discrepancy should remain. If the sulphide-lead was introduced, then it may have come from deeper parts of the granitic mass or from a subgranitic source. In the first case it is still granitic lead and again the atomic weight discrepancy should remain. In the second case a granitic origin for the sulphide-lead is denied.

Radioactivity and the source of Ore-lead.

Graton next attacks the hypothesis that the lead of lead ores has been derived from a source beneath the granitic and basaltic layers of the crust. He attempts to show that the hypothesis is

[17] Korzhinsky, D. S.: Mobility and inertness of components in metasomatosis. Bull. Acad. Sci. U. R. S. S., Cl. Sci. Math. Nat., Ser. Geol., 1936, pp. 35–57 (Russian), pp. 58–60 (English).

[18] Wells, R. C.: The origin of primary lead ores. ECON. GEOL., 33: 216, 1938.

false because it involves an internal contradiction. Graton supposes that the hypothesis implies a non-radioactive source for lead ores, and states that this implication is contradicted by the occasional association of radioactive ores with lead ores (*e.g.* pitchblende with galena at Jáchymov and Great Bear Lake). He confronts me (p. 280) with the italicized assertion that "*The mysterious depths are no more capable than the known magmas of yielding ore lead.*" My reply is simple. It is not the hypothesis that is contradicted, but the supposition that the source of lead ores is non-radioactive.

Graton states (p. 262) that "it seems to follow inescapably" from my argument that "the source from which ore lead has been at different times derived (to yield ores of different ages) has from the beginning been devoid of elements capable of yielding radiogenic lead." Again, on p. 280, he claims that I am forced into the *ad hoc* assumption that "the imagined deeper-lying material has never contained radioactive material capable of generating lead." Keevil also holds me responsible for the "fundamental assumption that somewhere in the earth there is a source free from radioactivity." I cannot agree that anything I have written implies a non-radioactive substratum or other source for ore-lead, and Graton himself admits that I have not mentioned this idea. I was, indeed, careful to point out (1937, p. 778) that if ore-lead be a derivative from ultrabasic rocks or magmas, its average atomic weight should vary—on the present limited data—from 207.20 to 207.21. But the ultrabasic rocks *are* radioactive and, since they cannot be ruled out by present atomic weight evidence as a potential source of ore-lead, I fail to see why Graton and Keevil should have gathered that I tacitly assumed the real but unknown source of ore-lead to be necessarily less radioactive than known ultrabasic rocks.

For well known thermal reasons it has generally been regarded as probable that the substratum is either devoid of, or very poor in, radioactive elements. I have personally always subscribed to the second alternative. The new radioactive determinations by Keevil and his colleagues strongly suggest that the substratum may be considerably more radioactive than has hitherto been sup-

posed possible, since the thermal difficulties implied by a relatively strongly radioactive crust disappear in the light of the new results. Moreover, the difficulty of accounting for the energy liberated from radioactive elements in the substratum may be further eased by the implications of recent investigations [19] which suggest that part of the matter of the substratum may be in the degenerate state. If degenerate matter be present, then the radiogenic energy liberated by disintegration may be largely utilized in raising such matter to the familiar non-degenerate state. I am indebted to Dr. D. S. Kothari for confirming the physical possibility of this inference.

The fact that the atomic weight of ore-lead ranges between 207.20 and 207.22 obviously leaves room for contamination of primeval lead by radiogenic lead, and even for a fair amount of such contamination if the radiogenic lead formed in the source material should itself have a resultant atomic weight near 207.21. The real point at issue is not that ore-lead has remained uncontaminated throughout geological time, but that the range of the effect of radiogenic lead on the atomic weight of ore-lead has been numerically small compared with the range of the effect of radiogenic lead on granitic or basaltic rock-lead.

That ore-lead has probably been more or less contaminated by radiogenic lead is strongly suggested by Nier's important discovery [20] that the component isotopes of different samples of ore-lead vary in relative abundance, despite the approximate constancy in atomic weight. Nier infers from his results that "the conclusions of Holmes may have to be modified, inasmuch as he used the alleged constancy of common lead as an argument against derivation of lead ores from acid or basic magmas." Commenting on this, Graton (p. 273) says, "this one lot of new quantitative determinations would seem sufficient to rupture Holmes' chain of argument."

[19] Kothari, D. S. and Majumdar, R. C.: Quantum statistics and the internal constitution of the planets. Nature, 137: 157–158, 1936. Proc. Nat. Acad. Sci. India, 6: 57–65, 1936.

[20] Nier, A. O.: Variations in the relative abundances of the lead isotopes. Am. Phys. Soc. Bull., 13: 17, 1938.

In reply to Nier, I need only point out that my conclusion was not based on the "alleged constancy of common lead," but on the approximate constancy of the atomic weight of common lead, which is a very different thing, since it allows for isotopic variation. Figs. 1 and 2 illustrate the contrast between this approximate constancy and the wide range of atomic weight of granitic rock-lead, as computed from radioactive data.

In reply to Graton I need only summarize the reasons for concluding that my argument remains undisturbed by the isotopic variation of ore-lead. If the cause of variation is radiogenic contamination—as, with Nier, I think most likely—this only means that the source-material of ore-lead also contains U and Th, which I have never doubted. Graton suggests (p. 272) that initial variations in the constitution of the earth's primeval lead may be, in part, the explanation of Nier's results. If so, then, as already stated, the only effect on the fans of Figs. 1 and 2 is that the point *A* becomes a vertical line representing the initial variation of atomic weight. Graton's other suggestion—that isotopic fractionation may be the explanation or contribute to it —has been discussed in the foregoing section, where it is dismissed as being without practical bearing on the point at issue.

It remains to be added that Nier's masterly attack on the problem is of critical importance and will probably go far towards finally solving it. Even though particular samples of rock-lead may be indistinguishable from ore-lead by atomic weight determination, the isotopic constitutions of the two are likely to be sufficiently different to be readily distinguishable by mass-spectrograph and other methods of isotopic analysis.

FURTHER PETROGENETIC CONSIDERATIONS.

Granitic and Basaltic Magmas.

On p. 276 of his paper, Graton suggests that a very important petrogenetic corollary follows from my inference that the average atomic weight of basaltic lead is different from that of granitic lead: the corollary being that "*granitic and basaltic magmas must have remained genetically independent since the earth-begin-*

ning!" He views this possibility with consternation and remarks that, if the corollary be correct, "our present-day hypotheses of magmatic differentiation in the broadest sense, our entire concept of petrogenesis, would be seriously embarrassed or rendered suspect." With regard to this remark I would direct attention to the fact that the 'concept of petrogenesis" to which Graton evidently refers has already been far more seriously embarrassed by purely petrological discoveries. The steadily expanding body of evidence recorded in the contributions previously listed, affords a spectacular proof that nothing less than a petrological revolution is now in full swing. There is already a great deal of evidence that many granitic rocks owe their origin to processes of metasomatism, rheomorphism (which includes magmatisation) and assimilation, whereas the view that granites are differentiates from basaltic magma is supported only by unsystematic association and belief in theoretical possibilities. Thus, the revolutionaries will not regard the atomic weight evidence with horror if it implies that granitic magma is not a residuum from basaltic magma.

The corollary italicized by Graton is worded somewhat more sweepingly than can be rigorously justified on the atomic weight evidence available at the moment. It seems to me fair to conclude from the present state of the data that the average atomic weight of basaltic lead is likely to be appreciably lower than that of granitic lead, and the range of individual values considerably wider. The probability that ore-lead cannot be basaltic lead is very high. Thus, if it is believed that ore-lead comes from granitic sources, it must at the same time be regarded as improbable that granitic magma is a residuum from basaltic magma. If it is not believed that ore-lead comes from granitic sources then—on this line of evidence—the independence of granitic and basaltic magmas could be inferred only from the following considerations. Figs. 1 and 2 show examples of the atomic weight fan for granitic rocks. The area of the corresponding fan for basaltic rocks is about the same as that of Fig. 1 and considerably greater than that of Fig. 2; it thus includes within itself the area of the granitic fan. It follows that corresponding to any granitic

sample containing rock-lead that had a given atomic weight at the time of consolidation there may be a basaltic sample containing rock-lead that had the same atomic weight at the time of consolidation. If, by a remarkable convergence of coincidences, the respective members of each such correlated pair also turned out to be members of the same igneous complex, the validity of the italicized corollary would be gravely weakened. But the likelihood that the latter condition should hold—apart from occasional inevitable coincidence—may reasonably be regarded as extremely improbable.

The doubt, however, could be resolved experimentally. The method would be to prepare samples of rock-lead (uncontaminated by introductions of ore-lead) from associated granitic and basaltic rocks of the same petrogenetic cycle, and then to determine the atomic weights and isotopic constitutions of the lead samples; determination of U and Th would also be necessary in the case of old rocks, to allow for the effect of radiogenic lead generated after the consolidation of the two rocks.[21] If the atomic weights (corresponding to the time of consolidation) were found to differ by more than the experimental error, or if the isotopic constitutions were found to be different, then it could be safely concluded that that particular granitic rock was not a differentiate from the magma represented by that particular basaltic rock.

The Relation of Lead Ores to Granitic Rocks.

Graton approves my statement that there is no evidence to suggest any direct genetic connection between lead ores and peridotite. Later, p. 278, he quotes my reference to Gregory's opinion that the common association of ores with igneous rocks is due to "the fact that intrusions rupture the crust and so pro-

[21] A few years ago I suggested a similar method of petrogenetic research, based on the ratio Ca^{41}/Ca in igneous rocks (Geol. Mag., 69: 550, 1932). This method has already been actively explored in collaboration with Dr. F. Allison, but publication of the results is being deferred, since it has become doubtful whether Ca^{41} is one of the disintegration products of potassium.

vide fractures and fissures through which the ore solutions can readily ascend." He then asks:

> If this mechanism can be depended on for granitic intrusives supposed to originate above the ore-source, does it not seem surprising that intrusions of peridotite from (or at least from nearer) the ore-source should be so rarely associated with ores as an earlier quotation from Holmes admits?
>
> And by what process shall we imagine these deep "ore solutions" to have separated from the peridotitic or other stuff of the deep ore-source?

If I accept the challenge presented by these questions, and so enter a dangerous realm of speculation where the guiding facts are few, it is only to suggest that answers may not be impossible. I cannot pretend to offer more than slight clues that may later become worthy of further development as relevant data accumulate.

To begin with, I am by no means sure that it is safe to suppose that granite intrusives altogether "originate above the ore-source," at least as regards the material of the granites. If granites originate by the rheomorphic action of alkaline emanations on pre-existing crustal rocks, it may be that the granite-making emanations themselves originate and ascend from deep sources. It is at least conceivable that the emanations are derived from a source nearer the ore-source than peridotites. There is evidence from rocks like kimberlite, biotite-pyroxenite and katungite that similar alkaline emanations have ascended from below a possible peridotite source.[22] It may also be noted here that Spurr [23] has suggested that the ores of Missouri—lead, zinc, copper, cobalt, nickel, barite and fluorite—are connected with invasion of a widespread (mica-) peridotite magma. Fluorite is a common associate of lead ores and Lindgren [24] has recently discussed the marked concentration of fluorite in the alkaline province that stretches from British Columbia to Mexico. He also writes (p. 169): "The great fluorite deposits of Illinois

[22] Holmes, A. and Harwood, H. F.: The petrology of the volcanic area of Bufumbira. Geol. Surv. Uganda, Mem. 3, Part II: 249–258, 1936–37.

[23] Spurr, J. E.: The ore magmas. 1923, p. 402 and p. 412, footnote 10.

[24] Lindgren, W.: Differentiation and ore deposition, Cordilleran region of the United States. *In* Ore Deposits of the Western States: Lindgren Volume, 1933, pp. 151–180.

and Kentucky will perhaps be found to have similar alkaline affiliations, though only scant dikes of peridotite and similar basic rocks show at the surface." The dikes referred to by Lindgren are mica-peridotites.

Thus, it may be that lead ores are not associated with ordinary peridotites because the lead-carrying emanations (when these occur) follow alkaline emanations. When the latter react with peridotites, rocks such as mica-peridotite are generated before the lead ores appear in the same province. Much more commonly, however, reaction with sialic crustal rocks will occur, sediments and schists being granitized and older granites being rejuvenated by rheomorphic palingenesis. In these processes associated basic rocks tend to be assimilated or otherwise to lose their identity in the general reconstitution. The suggested hypothesis is therefore consistent with the fact that lead ores and their associates are far more characteristically found near granitic rocks.

Gregory's view, that the fractures through which ore-solutions ascend are crustal fractures due to the associated intrusions, is incomplete, since it leaves the equally essential sub-crustal fractures out of consideration. Moreover, earlier crustal fractures, as well as subcrustal fractures, may be necessary for the formation and emplacement of granitic intrusions. This view has been expressed very cogently by Schmitt [25] in the following paragraph:

> It seems likely that the primary factor in the localization and association of "stocks," contact pyrometasomatism and ores, is a deep-cutting break of one sort or another in the earth's crust; that the contact pyrometasomatic deposits are not derived from the "stocks" but that their association is the result of localization along mutual outlets to the earth's surface.

The dependence of granitic intrusions and ore deposits on pre-existing channels opened by tectonic movements has also been stressed by Billingsley and Locke.[26]

Recent developments in the study of deep-focus earthquakes

[25] Schmitt, H.: Structural associations of certain metalliferous deposits in Southwestern United States and Northern Mexico. A. I. M. E., Cont. No. 38: 23, 1933.

[26] Billingsley, P. and Locke, A.: Tectonic position of ore districts in the Rocky Mountain Region. A. I. M. E., Tech. Pub. No. 501: 12, 1933.

have revealed the reality of sub-crustal fractures.[27] Relative to the orogenic belt with which they are associated, the deep foci are found to lie on more or less irregular planes dipping inland from the axis of the belt. Gutenberg and Richter (p. 285) have reached the conclusion "that the causative mechanism of normal and deep earthquakes is of the same type and that shearing displacements occur down to depths of the order of 700 kilometers." The hypothesis of sub-crustal convection currents [28] may possibly account for such internal ruptures in material subject to plastic flow. Where a horizontal current turns down in the neighbourhood of a mountain root, shearing and snapping are most likely to occur in the region of maximum curvature of the flow lines, as in the case of a glacier passing round a bend. However, whatever the cause may be, there are evidently shear zones in the substratum, linked into related series that may lead up to the crustal fractures associated with mountain building. Passageways are thus made available from time to time through which mobile materials from various depths in the substratum may find their way into the crustal regions.

To suggest a process whereby the "ore solutions" separate from the material of the substratum is to enter a field of speculation where analogy is likely to be, if not wholly misleading, at least inadequate, as a guide. From innumerable observations of successive replacements in metalliferous deposits, it is inferred that the various constituents were introduced as part of a procession or flux of emanations of changing composition. It is further clear that as the members of the procession passed through the "crowd" represented by the material traversed, some of them joined the crowd, while members of the crowd took

[27] Gutenberg, B. and Richter, C. F.: Depth and geographic distribution of deep-focus earthquakes. Geol. Soc. Am. Bull., 49: 249–288, 1938. See also the following: Visser, S. W.: Some remarks on the deep-focus earthquakes in the International Seismological Summary. Second Paper. Gerlands Beitrage z. Geophysik. 48: 254–267, 1936. Leith, A. and Sharpe, J. A.: Deep-focus earthquakes and their geological significance. Jour. Geol., 44: 877–917, 1936.

[28] Holmes, A.: Radioactivity and earth movements. Trans. Geol. Soc. Glasgow. 18: 559–606, 1928–29. The thermal history of the earth. Jour. Wash. Acad. Sci., 23: 169–195, 1933.

their places in the procession and migrated to higher levels. For this reason it is difficult to recognize with certainty the primary constituents of the procession and to trace them back stage by stage to their original sources. In the case under discussion, however, there is already a considerable probability that ore-lead and some proportion of the alkalies are primary constituents.

Some of those who hold that ore-lead comes from granitic residua believe that it, and its immediate associates, separate as fractional distillates that appear in turn as the vapour pressures of the successive gas phases rise above the external pressures there obtaining. Shearing in the substratum is likely to promote mobilization of potential gas phases, to localize their concentration along bands of reduced pressure, and to facilitate their migration towards the crust. The composition of the gas phases may be expected to vary with the composition of the substratum adjoining the places where shear fractures develop. If certain parts were rich in ore-constituents and others poor, the puzzling and unsystematic irregularities of ore distribution would obviously be accounted for.[29]

The alkaline granite-making emanations may be liberated more rapidly, in greater quantities, and from higher levels of the substratum than the ore-making emanations. Moreover, they are likely to be more readily miscible with magma than the ore-making emanations, and to be capable of intergranular migration through rocks at temperatures higher than those at which ore-replacements occur. Thus, besides the probability that migration towards the crust is differential and from sources at very different levels, there is the further possibility that the ore solutions, whether gaseous or otherwise, may accumulate in local pools until fissures develop in the crust, for example, in a consolidating body of granite, and so provide channels along which successive distillates can proceed to the upper levels where deposition occurs. Granitic masses are particularly likely to act as foci of upward migration, since they have already heated the crust to high levels

[29] A somewhat similar suggestion has been made by De Lury, J. S.: Primordial segregation of metals. Jour. Geol., 46: 756–763, 1938.

and so, aided by the internal pressure of their own volatiles, provided the conditions for inevitable fracturing.

I must insist that the above is frankly speculative to an extent that would be inexcusable were it not that Graton has challenged me to give "reasonably satisfying" answers to his questions. Reasonable speculations are not, however, necessarily satisfying, as the real facts are likely to be far more remarkable and surprising than guesses controlled by our woefully limited knowledge.

CONCLUSIONS.

1. The data from which it was originally concluded that ore-lead is not a derivative from granitic or basaltic materials have been further examined and discussed. It is concluded that the adoption of Hevesy and Hobbie's analyses of the lead contents of rocks is justified and that its use does not invalidate the argument, particularly as there is evidence that the lead contents of granitic rocks do not depart far from the average. It is agreed that determinations of uranium in granitic rocks are uncertain and that the possibility must be faced that many of the values hitherto recorded may be over-estimates. The average value of the ratio Th/U may therefore be correspondingly higher than that indicated by existing data. There is, however, no reason to suspect that all the errors in individual determinations have been so extreme and so peculiarly varied as to make the true values of Th/U always fall near 5 or 6. The range of Th/U in granitic rocks, even in the light of the most recent determinations, remains significantly considerable. Consequently, the atomic weights of granitic rock-lead (*i.e.* the sum of original plus radiogenic lead) should vary through a considerable range. If ore-lead be a concentrated derivative from granitic lead, then the atomic weights of different samples of ore-lead should show a similar variation, especially when the samples are of Hercynian or later age. Since this expectation is not realized it becomes probable that ore-lead is not derived from granitic sources. For basaltic rock-lead the calculated range of atomic weights is likely to be greater than for granitic rock-lead. It is therefore still more probable that ore-lead is not derived from basaltic sources.

2. The argument is not affected by the possibility that the atomic weight of the earth's primeval lead may not have been everywhere the same, since any such variation could not appreciably reduce the range of the atomic weight of rock-lead and would be more likely to increase it.

3. The range of the atomic weight of granitic rock-lead appears to overlap that of ore-lead. That is to say, some granites may contain rock-lead having atomic weights indistinguishable from those of ore-lead. These granites might theoretically yield ore-lead, but, since differentiation cannot depend on a slight variation in atomic weight, there is no reason why they should be supposed to have done so, unless they happened to be abnormally rich in primary lead. There is no evidence that such primary-lead-rich granites exist.

4. The suggestion that selective extraction of lead isotopes from rock-lead might provide an extract of ore-lead of the right atomic weight is dismissed. Such a process would concentrate the lighter isotopes in the extract, if it occurred at all. Actually, however, the process does not appear to operate under natural conditions.

5. There is no reason to postulate a non-radioactive source for ore-lead. Isotopic analyses suggest that the varied isotopic constitution of ore-lead is due to contamination by radiogenic lead. The fact that the effect of such contamination on the atomic weight of ore-lead has been very slight indicates, in the light of existing data, that the source of contamination was not usually granitic or basaltic.

6. The recorded value of the atomic weight of cotunnite-lead from Vesuvius having been found unacceptable, the prediction that the atomic weight of rock-lead should usually be different from that of ore-lead is at present without direct experimental confirmation. It is understood that plans are now well under way for intensive investigation of carefully selected samples of indubitable rock-lead.

7. If it is believed that lead ores are derivatives from granitic magma, then it is practically impossible to believe at the same time

that granitic magma is a differentiate from basaltic magma. If it is not believed that lead ores are granitic derivatives, then the data leave room for some uncertainty, but suggest the improbability that granitic magma can be a differentiate from basaltic magma.

8. The suggestion that ore-lead is a derivative from deep-seated peridotite, *via* the successive differentiates of a single magmatic cycle from basaltic to granitic magma and thence to ore-solutions, is regarded as untenable on purely geological and petrological grounds.

9. It is suggested that ore-lead may be derived directly from lead-rich portions of the substratum when these are tapped by shear-fractures of the kind inferred from the records of deep-earthquakes.

UNIVERSITY OF DURHAM,
DURHAM, ENGLAND,
Sept. 6, 1938.

PYROSYNTHESIS, IDENTIFICATION AND STUDY OF TIN SULPHIDES AND OF COMPOUNDS OF TIN SULPHIDES WITH ANTIMONY AND LEAD SULPHIDES.

A. M. GAUDIN AND W. T. HAMLYN

ABSTRACT.

Tin is regarded as forming two sulphides, the monosulphide, which is known to mineralogists as herzenbergite, and the disulphide which is mineralogically unknown but commercially utilized as "mosaic gold." Both of these sulphides have been synthesized in this study, as likewise a third sulphide of intermediate composition. This third sulphide or sesquisulphide is probably Sn_2S_3 or $SnS \cdot SnS_2$.

Heretofore, the system tin-antimony-sulphur had been investigated but in part, and inaccurately. This investigation has disclosed two compounds of SnS with Sb_2S_3, namely phase A, $SnS \cdot Sb_2S_3$ and phase B, $3\ SnS \cdot Sb_2S_3$. No compound was found of SnS_2 with Sb_2S_3, but a characteristic compound of SnS, SnS_2 and Sb_2S_3 was obtained. The formula of this phase is not perfectly settled, but is perhaps $4\ SnS \cdot Sn_2S_3 \cdot Sb_2S_3$. No natural minerals have been described in this system.

Tin and lead sulphides are supposed to form one compound, teallite, whose formula is variously given as $PbS \cdot SnS$ and $PbS \cdot SnS_2$. This compound could not be synthesized. Instead it was found that SnS and Sn_2S_3 dissolve in solid solution large quantities of lead sulphide.

FILMING.

PROGRESS in this study would have been wholly impossible without the use of selective iridescent filming, but by that technique the various phases can be recognized readily. Caution is necessary, however, even in that method, since solid-solution effects of considerable magnitude, if not properly evaluated, lead to erroneous diagnoses.

The filming solutions previously set up as standards for the identification of other minerals were found unsatisfactory for filming the pure tin sulphides. But they were useful in many cases for the identification of complex sulphides of tin with other

metals. To that end two standard solutions were used. These are the hydrochloric acid-chromic trioxide-water solution already described [8] and a corresponding solution of sulphuric acid, chromic trioxide and water.[18] For the differentiation of pure tin sulphides a new solution was devised consisting of silver nitrate in aqueous nitric acid. Although the best of the many solutions that were concocted for that purpose this solution leaves much to be desired.

The Standard Silver Nitrate-Nitric Acid Solution.—The solution finally adopted was a saturated solution of silver nitrate in 50 per cent nitric acid by volume. This solution will be termed the standard silver nitrate solution.

At room temperature (18–20°) the solution is slow and does not give uniform results. However, good differentiation is possible where the film is uniform. At somewhat higher temperatures (25–35°) the films are progressively better, more uniform, and form faster.

In order to compile filming data for various temperatures and to be able to reproduce results a thermostatically controlled water bath was devised for the solution. The thermostat could be set for any temperature between 0 degrees Centigrade and 60 degrees Centigrade.

Some work was done in an effort to determine the chemical nature of the films produced by this solution. The weight of evidence points to silver sulphate as the composition in most cases. The evidence and other considerations are presented below:

1. The films are slowly soluble in water—the resultant solution gives a positive test for silver ions.
2. The same is true for ammonium hydroxide solutions.
3. Fifty per cent nitric acid dissolves the film rapidly—the resultant solution gives a positive test for silver ions.
4. The solution produces films on all sulphides tried to date.
5. Silver ions will replace any metal in a sulphide if the solubility of silver sulphide is less than that of the other metal sulphide but the rate is not predictable.
6. Silver sulphide is easily oxidized to silver sulphate by strong nitric acid.[9]
7. Following the reasoning of the mass action law as applied to solu-

bilities, silver sulphate should be relatively insoluble in a solution having such a high concentration of silver ions.

In view of the above evidence it is probable that the standard silver nitrate solution differs from other solutions used in selective iridescent filming in that it requires replacement of metal ions before a film is produced.

In reviewing the possibilities of this solution, it is well to state that this solution is not recommended for use except with refractory sulphides that do not respond to other oxidizing solutions. The sulphides of tin belong to this class of compounds.

Other Solutions Tried on Tin Sulphides.—The solutions and results obtained are: HNO_3-MeOH, poor results—etches and films; H_2O_2-H_2O, negative; CrO_3-H_2O, negative; I_2-MeOH, etching; $AgNO_3$-H_2O, replacement; H_2SO_4-MeOH, etching; $KClO_4$-H_2SO_4-H_2O, some etching; H_2SO_4-CrO_3-H_2O very slow —some etching; HCl-CrO_3-H_2O, etching; H_3PO_4-HNO_3-H_2O, very slow; HCl-HNO_3-H_2SO_4-H_2O, etching; $AgNO_3$-HNO_3-H_2SO_4-H_2O, very slow; $CuSO_4$-HNO_3-H_2O, slow—etching; $Zn(NO_3)_2$-HNO_3-H_2O, negative; I_2-H_2SO_4-MeOH, slow—non-adherent films; I_2-H_2SO_4-HCl-MeOH, non-adherent films; I_2-HCl-MeOH, too fast—non-adherent films.

The solution named last was used to some extent in studies of the tin-lead system. The method of synthesis has already been described.[10]

THE SYSTEM TIN-SULPHUR.

Chemical literature [9] records no complete equilibrium diagram of the system tin-sulphur. Investigators have completed the diagram to just beyond the first sulphide of tin on the high tin side of the diagram. Investigations of high-sulphur charges have been unsuccessful due to the high vapor pressure of tin disulphide. The literature records two definitely established sulphides of tin, SnS and SnS_2. A third, Sn_2S_3 to which reference is made by Berzelius is generally discredited by chemists.[9, 12] Mineralogists have recently included the monosulphide, SnS, as a natural mineral, herzenbergite.[11] Table I includes the prin-

cipal known characteristics of the phases observed. Tables II and III present the filming characteristics of the various phases and phase-variations. Photomicrographs show the principal structures observed.

TABLE I.

PROPERTIES OF THE PHASES.

Phase	Composition	Probable Crystal System	Polarization	Natural Color	Color under Vertical Illumination	Polishing Ability	Transmission of Light
Herzenbergite	SnS	Orthorhombic	Anisotropic	Light grey	Very light grey	Medium	Opaque
Sn_2S_3	Sn_2S_3	Orthorhombic	Anisotropic	Grey	Mouse grey	Good	Slightly translucent
SnS_2	SnS_2	Hexagonal	Anisotropic	Reddish-orange	Mouse grey to dark grey	Poor	Translucent
Galena	PbS	Isometric	Isotropic	Grey	Light grey	Good	Opaque
Stibnite	Sb_2S_3	Orthorhombic	Anisotropic	Lead grey	Grey	Good	Opaque
Phase A	$SnS \cdot Sb_2S_3$	Orthorhombic	Anisotropic	Lead grey	Grey	Good	Opaque
Phase B	$3SnS \cdot Sb_2S_3$	Orthorhombic	Anisotropic	Lead grey	Grey	Fair	Opaque
Phase C	$4SnS \cdot Sn_2S_3 \cdot Sb_2S_3$ (?)	Isometric	Isotropic	Light grey	Very light grey	Good	Opaque

TABLE II.

IDENTIFICATION GUIDE NO. 1

STANDARD SILVER NITRATE SOLUTION (25° C.).

Sulphide	Time of Immersion					
	15 Seconds	30 Seconds	1 Minute	2 Minutes	5 Minutes	10 Minutes
SnS	Yellow (I)	Orange-brown to violet (I)	Blue (I) to yellow (II)	2nd order colors		
Sn_2S_3		Light salmon (I)	Violet-brown (I)	Violet-brown to violet (I)	Violet to blue (I)	Blue-violet to blue (I)
SnS_2					Dark yellow-brown (I)	Yellow-brown to yellow (I)

Note: Numbers in parenthesis refer to the order of the colors.

TABLE III.

IDENTIFICATION GUIDE NO. 2.

STANDARD HYDROCHLORIC ACID-CHROMATE SOLUTION (18–20° C.).

Phase	Time									
	5 Seconds	10 Seconds	15 Seconds	20 Seconds	25 Seconds	30 Seconds	1 Minute	2 Minutes	5 Minutes	10 Minutes
Stibnite	Pale yellow	Yellow	Yellow-brown to violet-brown	Violet-brown to violet	Blue-violet to blue	Light blue	Reddish violet (II)			
Lead-bearing* stibnite	Faint yellow	Yellow	Yellow-brown to violet-brown	Violet-brown to blue-violet	Blue to yellow (II)					
Phase A		Pale yellow	Pale yellow	Yellow	Yellow to yellow-brown	Yellow-brown to violet-brown	Blue			
Lead-bearing* phase A	Very pale yellow	Pale yellow	Yellow	Yellow-brown	Violet-brown	Violet	Yellow (II)			
Phase B					Pale yellow	Light yellow	Violet-brown to violet	Blue		
Lead-bearing* phase B					Light yellow	Yellow	Violet to blue-violet	Yellow (II)		
Herzenbergite	Unfilmed by this solution									
Antimony-bearing* herzenbergite								Very pale yellow	Deep yellow-brown	Blue to yellow (II)
Lead-bearing herzenbergite*	Unfilmed by this solution but etched									
Phase C							Very pale yellow	Yellow-brown	Violet-brown to blue-violet	
Tin sesquisulfide	Unfilmed by this solution									
Antimony-bearing* sesquisulfide								Very pale yellow	Deep yellow brown	Blue to yellow (II)
Lead-bearing* sesquisulfide	Unfilmed by this solution									
Tin disulfide	Unfilmed by this solution									

Note: Numbers in parenthesis refer to the order of the colors. First order colors are not so designated.

* Data marked thus have been obtained in connection with a study of the system lead-tin-antimony-sulphur which is not yet completed

Lead-bearing* sesquisulfide	Unfilmed by this solution
Tin disulfide	Unfilmed by this solution

Note: Numbers in parenthesis refer to the order of the colors. First order colors are not so designated.
* Data marked thus have been obtained in connection with a study of the system lead-tin-antimony-sulphur which is not yet completed

Discussion of the Structures and Phases.

Tin (*Sn*).—In the specimens low in sulphur, metallic tin occurs in a very fine grained intergrowth with monosulphide. This intergrowth contains less than 2 per cent of monosulphide and occurs as round globules embedded in the sulphide. From this structure it is concluded that metallic tin and tin sulphide are but slightly soluble in the liquid state.

The fine intergrowth of sulphide in metal is easily differentiated from the monosulphide by its cream color and by its relief when polished. When viewed at low magnification it resembles metallic tin. If the specimen is given a dry buffing the tin assumes a burnished appearance and brown color whereas the monosulphide is not affected (Fig. 1).

All the filming solutions etch metallic tin.

Tin Monosulphide (*SnS*), (*Herzenbergite*).—With the tin-sulphide intergrowth the monosulphide is massive or occurs as bright grey needles embedded in it. With the sesquisulphide the monosulphide occurs as primary brilliant light-grey needles in eutectic intergrowth (Fig. 2). It does not occur in contact with the disulphide.

The monosulphide is easily differentiated from the other sulphides by its lighter color, greater hardness, extreme brilliance and structure. It is anisotropic.

The standard silver nitrate solution produces fair films quite rapidly on the monosulphide—a good first order violet in 30 seconds (Table II). The chromate solutions etch this sulphide without producing a film.

Tin Sesquisulphide (Sn_2S_3).—This sulphide occurs as a later phase surrounding the monosulphide and in eutectic relation with it. It crystallizes at a lower temperature than the disulphide; this is shown by specimens in which it surrounds the disulphide. No eutectic relation was observed between the sesquisulphide and the disulphide.

The sesquisulphide is a mouse-grey anisotropic phase showing some internal reflections. It probably belongs to the orthorhombic system as shown on specimens where it appears as lozenge-shaped crystal sections.

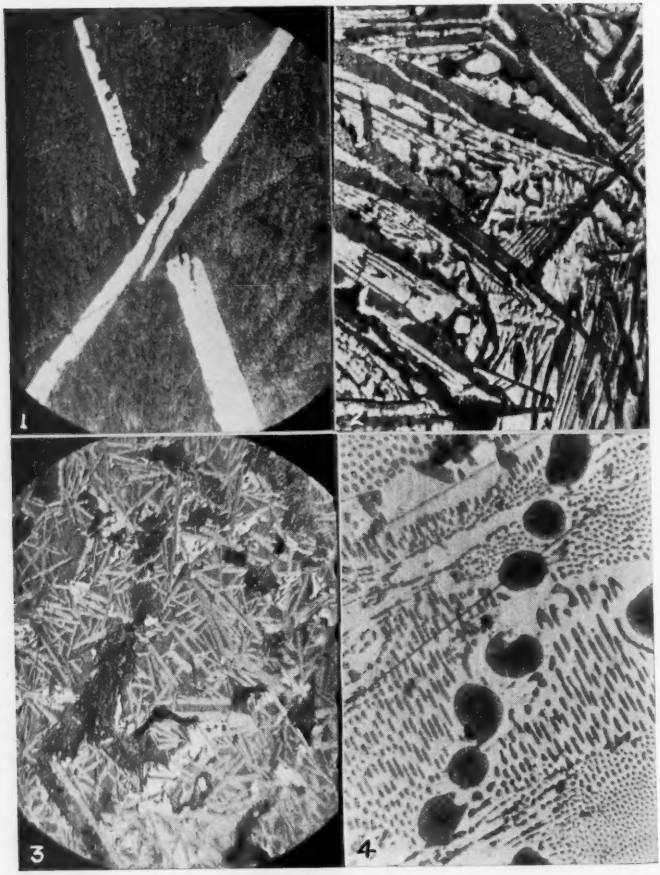

FIG. 1. Needles of tin monosulphide (white) in a groundmass of fine grained sulphide-metal eutectic. Unfilmed; metal buffed brown. × 140, air.

FIG. 2. Primary needles of tin monosulphide, blue (dark grey) in an eutectic of monosulphide and sesquisulphide, violet-brown (white). Filmed for one minute in the Standard Silver Nitrate Solution. × 135, air.

Some analyses were made in an effort to determine the true chemical composition of this phase. The specimens used for these assays were specimens having a considerable amount of lead sulphide. The lead sulphide apparently complicated the procedure to such an extent that the assays were not accurate. However, the assays do indicate that the sulphide is intermediate between the monosulphide and the disulphide, thus supporting microscope evidence. The composition is given as that of a sesquisulphide, the most logical compound from a chemical view point but this is not yet fully supported by chemical analyses.

The standard silver nitrate solution produces fair films slowly. A first order mauve-brown is obtained in two minutes. The standard sulphuric-chromate solution gives a poor film accompanied by etching.

Tin Disulphide (SnS_2).—The disulphide occurs as a border phase between the lower sulphides and sulphur in some high-sulphur specimens, as incrustations on the top of others and as plates embedded in sulphur (Fig. 3). It is also present as a sublimate on the walls of crucibles and on the walls and top of bombs in high-sulphur specimens.

By reflected light the disulphide appears as a dark-grey anisotropic phase occurring as plates that display brilliant orange internal reflections with crossed nicols under oil except where the plates are cut on end. It decomposes at some temperature between 575° and 600° C. This was determined by chilling high-sulphur specimens at definite temperatures—the growth of the disulphide was used as an index. The specimen chilled at 575° showed a marked increase in the amount of disulphide present over those chilled at a higher temperature.

The disulphide apparently crystallizes from either the vapor

FIG. 3. Plates of tin disulphide (light grey) in irregular areas of sesquisulphide (white). Sulphur (medium grey) filling in around plates of tin disulphide. Grey areas are pits. Unfilmed. × 140.

FIG. 4. Dendritic blobs of galena in a groundmass of galena—sesquisulphide eutectic structurally controlled by the sesquisulphide. Filmed one sec. in the Standard Hydrochloric Acid-Chromate Solution. Colors: galena blue (grey); sesquisulphide, light grey (white). × 275.

phase or the liquid phase. At first, it was believed that it formed only from the vapor phase; later studies of some specimens indicate that it also solidified from the liquid state.

This sulphide is very slowly filmed by the standard silver nitrate solution—it acquires a first order yellow in 10 minutes.

Sulphur (*S*).—In high-sulphur specimens, sulphur occurs as a cap on the specimens or as small inclusions in the specimens.

It is readily distinguished from the other phases by its yellow color under oil or under crossed nicols. A good test for it consists in placing a small drop of CS_2 or some other solvent for sulphur on the phase suspected of being sulphur—if it is sulphur a deep etch will be observed.

Conclusions.

Tin forms three distinct sulphides, SnS, Sn_2S_3 and SnS_2. Sn and SnS are but partly soluble in the liquid state. A eutectic occurs between SnS and Sn_2S_3; no such relation was found between Sn_2S_3 and SnS; Sn_2S_3 appears to have an incongruent melting point. Sulphur is always the last phase to solidify. The disulphide apparently decomposes between 575° and 625° C. The sketch of the system (Fig. 5), is, therefore, not a true equilibrium diagram since the sulphur vapor pressure was not kept constant but allowed to vary with the temperature. The curves are only approximate.

THE SYSTEM TIN-LEAD-SULPHUR.

This system has been treated until now as a binary system, whereas in reality it is a ternary system. A portion of the system involving SnS and PbS has been worked out and no compound reported.[13] A eutectic containing approximately 45 per cent PbS by weight is reported .

Mineralogists list one compound in this system, teallite. They are however, not in agreement as to its composition—one group of mineralogists give its composition as $PbS \cdot SnS$ [3, 14, 15] while the other list it as $PbS \cdot SnS_2$.[4]

No success was had in the many attempts to synthesize teallite.

At first it was believed that teallite had been synthesized but it was later realized that what had been taken for teallite was tin monosulphide containing large amounts of lead sulphide in solid solution.

Discussion of the Structures and Phases.

Tin Monosulphide ((*Sn, Pb*)*S*).—This sulphide of tin was found in all the low-sulphur specimens. It holds large amounts of lead sulphide in solid solution.

This phase is found in a eutectic-like relation with galena and with the sesquisulphide of tin. When primary it occurs in a

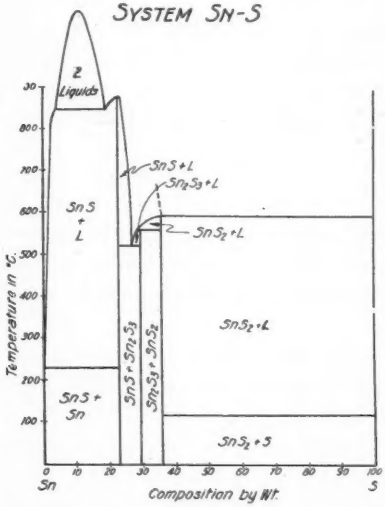

FIG. 5. System Sn–S.

thick, blunt habit instead of in the acicular habit characteristic of pure herzenbergite. Gradations in habit are also observed.

The rate of filming for pure tin monosulphide and pure lead sulphide by the standard silver nitrate solution being about the same, this solution cannot be used to differentiate between them. The standard chromate-hydrochloric acid solution films tin monosulphide slowly and with a slight etch.

Tin Sesquisulphide ($Sn,Pb)S \cdot SnS_2$).—This sulphide is found both primary and in a eutectic-like relation with the monosulphide and galena (Fig. 4). It apparently dissolves a considerable amount of lead sulphide although less than the monosulphide (Fig. 8).

The sesquisulphide with lead in solution is filmed proportionately faster by the standard silver nitrate solution. The hydrochloric acid-chromate solution also films lead-bearing sesquisulphide at a slow rate and in accordance with the amount of lead sulphide in solution (Table III).

The other properties of this sulphide have already been described.

Tin Disulphide (SnS_2).—This sulphide occurs in isolated patches with sesquisulphide and bears no relation to the other phases. It apparently does not take up any lead sulphide in solid solution as its rate of filming in the standard silver nitrate is not affected.

Galena (PbS).—Galena bears a eutectic-like relation to the sesquisulphide and to the monosulphide. In the high-lead specimens it occurs as rounded blebs that are primary. The sulphide must hold some tin sulphide in solid solution as it is filmed slightly slower than pure galena by the hydrochloric acid-chromate solution.

The standard chromate solutions film this phase very rapidly. The standard silver nitrate solution is much slower (Blue I in 1 min.).

Conclusions.

At temperatures of the order of magnitude of 500° C. or higher tin sulphides apparently form no compounds with lead sulphide. All the relations observed have been those of complete solubility in the liquid state and partial but large solubility in the solid state.

This relation between the monosulphide and lead sulphide is to be expected as the metals are closely related chemically in spite of the difference in crystal system of their sulphides. One would

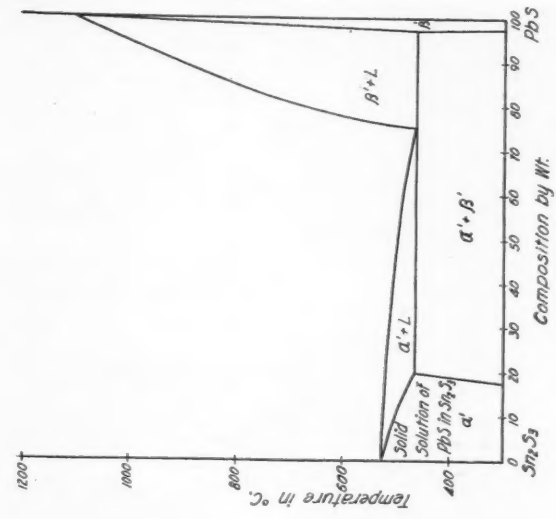

FIG. 6. System SnS—PbS.

FIG. 7. System Sn_2S_3—PbS.

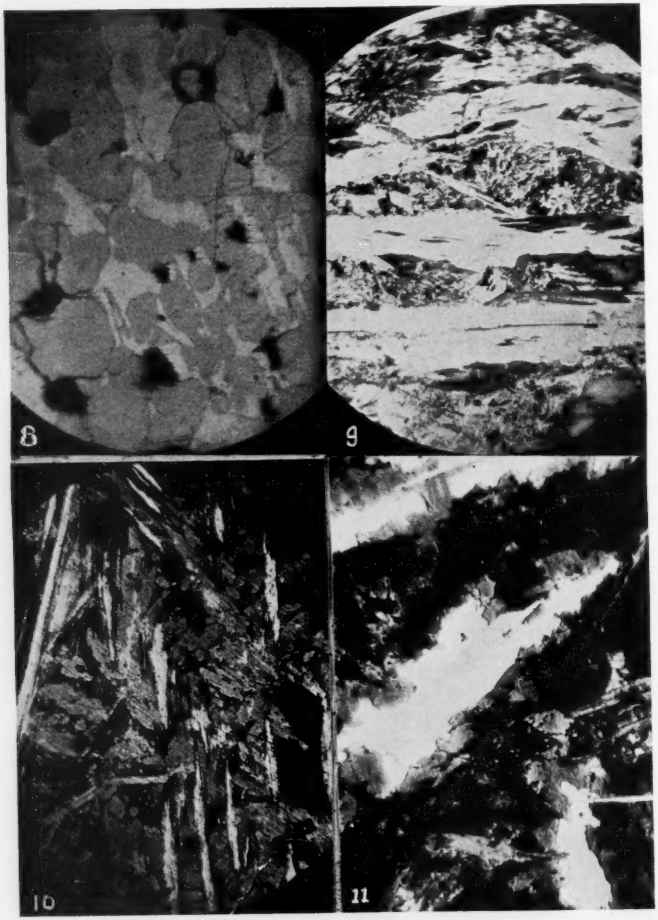

FIG. 8. Primary tin sesquisulphide (grey) surrounded by interstitial tin monosulphide (light grey). This specimen contains 90 per cent Sn, 10 per cent Pb, but no lead mineral is seen. Unfilmed; × 140, air.

FIG. 9. Elongated crystals of Phase A, deep yellow (white) in a eutectic groundmass of Phase A and stibnite blue (grey). Filmed 27 sec. in Standard Hydrochloric Acid-Chromate Solution. × 140.

expect a tendency to form solid solutions instead of chemical compounds. Compounds between lead sulphide and the other sulphides of tin would be more likely, chemically. However, none were found.

These relations are shown graphically in the pseudo-equilibrium diagrams (Figs. 6 and 7). Temperatures and compositions were determined in a very rough manner only.

THE SYSTEM TIN-ANTIMONY-SULPHUR.

In the system SnS-Sb_2S_3 chemists record [13] one compound only. This has an incongruent melting point and bears a eutectic relation to stibnite. Re-investigation of this system disclosed besides this compound (phase A) another low-valent tin-antimony sulphide (phase B), which has a higher tin content, also an incongruent melting point. Other than the present no studies are available of the tin-sulphur-antimony system with large quantities of sulphur. A compound (phase C) having tin in an intermediate valence was found—none was found having tin in the divalent state.

Mineralogists record no compounds in this system but it would not be too much to expect that they occur in nature.

Discussion of the Structures and Phases.

Stibnite (Sb_2S_3).—Stibnite occurs in all the specimens prepared for this system. It is found in eutectic-like structure with phases A (Fig. 9) and C, and with tin disulphide.

Stibnite is a light grey anisotropic phase with a tendency to crystallize as lozenge-like crystals when free to do so. Unfilmed

FIG. 10. Small blade-like crystals of phase B, pale yellow (white), surrounded by phase A, deep yellow (grey). Smaller crystals of phase A in a eutectic groundmass of phase A and stibnite, blue (dark grey). Filmed 27 sec. in Standard Hydrochloric Acid-Chromate Solution. × 140.

FIG. 11. Large irregular primary crystals of tin monosulphide, pale yellow (white), surrounded by phase B, violet brown (grey). Groundmass of phase A, blue (dark grey). Filmed 1 min. in Standard Hydrochloric Acid-Chromate Solution. × 140.

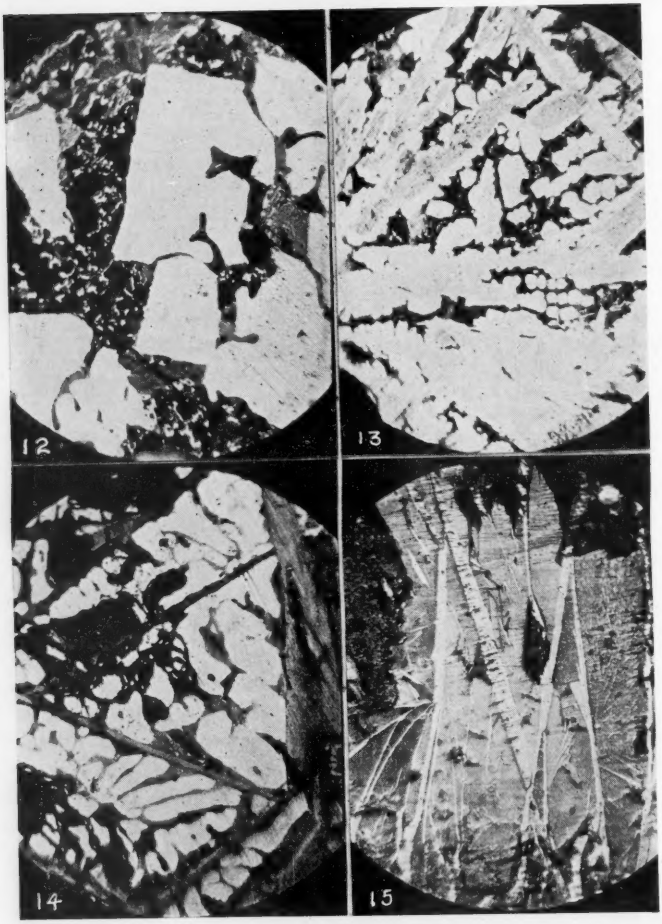

FIG. 12. Large regular crystals of phase C (white) in a eutectic groundmass of phase C and stibnite, blue (dark grey). Filmed 27 sec. in Standard Hydrochloric Acid-Chromate Solution. × 140.

FIG. 13. Regular needle-like and lozenge-like crystals of tin sesquisulphide (pale grey) surrounded by phase C (white). Groundmass is a

it cannot be distinguished from phase A, B or C as the color differences are negligible.

Stibnite acquires a characteristic blue color after filming for 27 seconds in the standard chromate-hydrochloric acid solution. The standard silver solution films stibnite slowly and the results are not reliable.

Phase A ($SnS \cdot Sb_2S_3$).—This phase is the one recorded in chemical literature. It occurs in the low sulphur specimens as a border phase around Phase B (Fig. 10) and as allotriomorphic crystals (Fig. 9). It occurs with stibnite as a eutectic. There is some solid solution between this phase and stibnite as areas of this phase around large crystals of stibnite film at a faster rate. In higher-sulphur specimens this phase is late and occurs with the stibnite. This phase is light grey and difficult to distinguish from stibnite. It is anisotropic. The standard chromate-hydrochloric acid solution films it to a yellow-brown I in 27 seconds. The composition of the phase was checked accurately by microscope counts.

Phase B ($3SnS \cdot Sb_2S_3$).—The composition of this phase has been deduced from microscope estimates; while it is believed correct, the accuracy is not as high as that with which phase A is known. Phase B occurs as a border phase around tin monosulphide (Fig. 11) or as long primary needles or patches in phase A (Fig. 10). It is light grey, anisotropic and one which in some orientations shows some relief against phase A. It acquires a pale yellow I color when exposed to the standard chromate-hydrochloric acid solution for 27 seconds.

Tin Monosulphide ($(Sn,Sb)S$).—This sulphide is the same as the one previously described except that it holds a fair amount of

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eutectic of phase C and stibnite, blue (dark grey). Filmed 27 sec. in Standard Hydrochloric Acid-Chromate Solution. × 140.

FIG. 14. Plates of primary tin disulphide (dark grey). Rounded crystals of phase C (white) embedded in a eutectic of phase C and stibnite, blue (dark grey). Filmed 27 sec. in the Standard Hydrochloric Acid-Chromate Solution. × 140.

FIG. 15. Plates of tin disulphide embedded in stibnite, yellow (grey) and in a fine-grained eutectic of the disulphide and stibnite. Filmed 15 sec. in Standard Hydrochloric Acid Chromate Solution. × 140.

antimony sulphide in solution (perhaps up to 8 per cent) and so is slowly filmed by the chromate solutions. It occurs as small blobs and large crystals surrounded by phase B (Fig. 11). In all specimens it stands out in bold relief from the other sulphides.

The standard chromate-hydrochloric acid solution films this sulphide a first order blue in 5 minutes. For comparison it may be remembered that pure tin monosulphide is not filmed by this solution. The stibnite in solid solution retards the filming rate in the standard silver nitrate solution remarkably—the results are spotty and unreliable.

Phase C ($4SnS \cdot Sn_2S_3 \cdot Sb_2S_3$).—From microscope estimates it seems that this phase contains three atoms of tin to one of antimony. The formula is perhaps $3Sn_2S_3 \cdot Sb_2S_3$, or $4SnS \cdot Sn_2S_3 \cdot Sb_2S_3$. It could not be ascertained accurately as specimens containing more than 50 to 60 per cent of phase C were not obtained.

This phase occurs in eutectic relation with stibnite and as large primary crystals showing good cubic outlines (Fig. 12). With tin disulphide or sesquisulphide it is secondary (Figs. 13 and 14) and with phase A, primary. It is difficult to obtain in specimens containing much antimony as the sulphur control becomes more difficult with increasing antimony. Phase C is brilliant white, isotropic, and shows little or no relief against stibnite. It is filmed blue in 5 minutes by the standard chromate-hydrochloric acid solution.

Tin Sesquisulphide ($(Sn,Sb)_2S_3$).—This phase also appears to hold stibnite in solid solution as it is slowly filmed by the chromate-hydrochloric acid solution.

This phase occurs as lozenge shaped crystals probably belonging to the orthorhombic system. In high-tin medium-sulphur specimens it forms cores inside of phase C (Fig. 13). It displays appreciable relief against all the other phases, except the monosulphide, which is harder.

Tin Disulphide (SnS_2).—This phase occurs with stibnite (Fig. 15), phases A and C, and the sesquisulphide, but not with phase B. It is primary with all. It may under the proper circum-

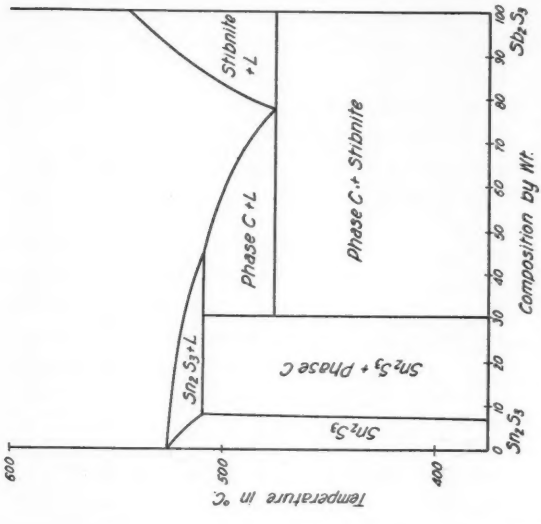

FIG. 17. System Sn_2S_3-Sb_2S_3.

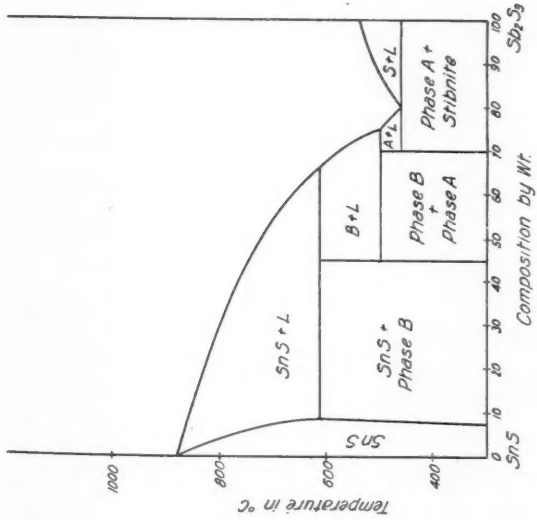

FIG. 16. System SnS-Sb_2S_3.

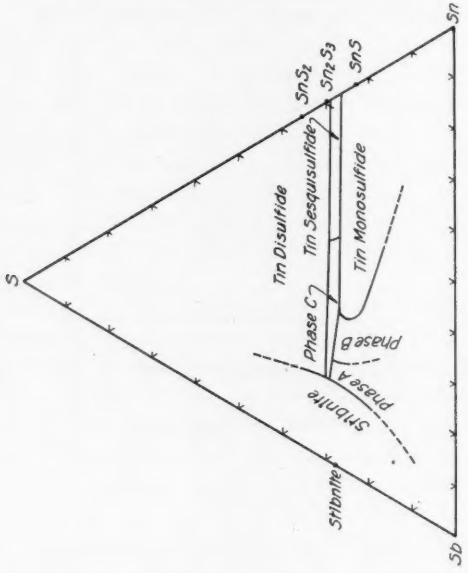

FIG. 19. System Sn-Sb-S.

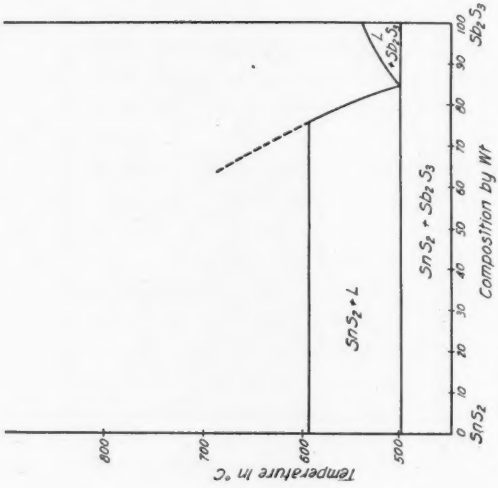

FIG. 18. System SnS_2-Sb_2S_3.

stances be secondary to stibnite, but such a relationship was not observed. It crystallizes as plates which in section appear as dark grey needles.

Under oil this phase displays reddish internal reflections with crossed nicols. In the presence of much stibnite that has been filmed blue this sulphide appears to have a flesh color, which is nonexistant if the stibnite is filmed to any other color than blue. This effect may be an optical illusion.

Sulphur (*S*).—Sulphur is present as the latest phase. When adjacent to it stibnite displays good crystal outlines. Sulphur may be confused with the disulphide but it is darker and lacks crystal outlines and shows pale yellow internal reflections when viewed under oil and with crossed nicols.

Conclusions

Tin monosulphide forms two compounds, A and B, with stibnite. Both have incongruent melting points. Tin sesquisulphide, or tin sesquisulphide and tin monosulphide together, form one compound with stibnite (C). This phase has an incongruent melting point and bears a eutectic relation to stibnite.

Tin disulphide forms no compounds with stibnite.

The results are shown by the pseudo-equilibrium diagrams which follow (Figs. 16 to 19).

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Aug. 26, 1938.

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16. JAEGER, AND VAN KLOOSTER: 2, Anorganishe und Allgemeine Chemie, 78: 245, quoted in Int. Crit. Tables, IV, 47.
17. IITUSKA: Memoirs of the College of Science, Kyoto Imp. Univ., 4: 61, quoted in Int. Crit. Tables, IV, 47.
18. GAUDIN, A. M., AND DICKE, GÜNTHER: The pyrosynthesis, microscope study and iridescent filming of sulphide compounds of copper with arsenic, antimony and bismuth. In press.

THE SIGNIFICANCE AND NATURE OF THE CONE OF DEPRESSION IN GROUND-WATER BODIES.[1]

CHARLES V. THEIS.

ABSTRACT.

In nature the hydraulic system in an aquifer is in balance; the discharge is equal to the recharge and the water table or other piezometric surface is more or less fixed in position. Discharge by wells is a new discharge superimposed on the previous system. Before a new equilibrium can be established water levels must fall throughout the aquifer to an extent sufficient to reduce the natural discharge or increase the recharge by an amount equal to the amount discharged by the well. Until this new equilibrium is established water must be withdrawn from storage in the aquifer and conversely the new equilibrium cannot be established until an amount of water is withdrawn from storage by the well sufficient to depress the piezometric surface enough to change the recharge or natural discharge the proper amount. The depression of the piezometric surface is called the cone of depression.

The characteristics of the cone of depression in an idealized aquifer of infinite areal extent are considered mathematically. Time is an essential variable in the description of this cone. The time rate of lateral growth of the cone is independent of the rate of discharge by the well and depends only on the physical characteristics of the aquifer. The cone must grow laterally much more rapidly in artesian aquifers than it does in nonartesian.

The characteristics of the cone in actual aquifers are then considered. In finely porous artesian aquifers the cone appears to be much like the cone in the ideal aquifer. The cone in nonartesian aquifers must be somewhat warped. The theory outlined implies that well discharge from extensive water-table aquifers must be a draft on storage in the aquifer for many generations.

ACKNOWLEDGMENTS.

This paper is in many ways an attempt to extend and refine the concepts pioneered by O. E. Meinzer regarding safe yield of aquifers in general and the compressibility of artesian aquifers.

[1] Published with the permission of the Director of the United States Geological Survey, and presented before the Society of Economic Geologists, Washington Meeting, Dec. 28, 1937.

The debt to his work is too general to be acknowledged specifically in all respects. The writer is also greatly indebted to his colleagues of the Division of Ground Water of the Geological Survey for many helpful discussions and for data bearing on the problem. Finally acknowledgment is due T. M. McClure, State Engineer of New Mexico, in financial cooperation with whose office most of the work was done.

THE SIGNIFICANCE OF THE CONE OF DEPRESSION.

WHEN a well discharges by pumping or artesian flow, water levels in wells in its vicinity are lowered. The lowering of the water table or other piezometric surface thus represented has been called the cone of depression or the cone of influence of the well. Much of the literature of ground water has tended to minimize the extent of the cone of depression and to consider it a local phenomenon. Thus Slichter,[2] Turneure and Russell,[3] and Tolman[4] give formulas involving the radius of the cone of depression, or "area" or "circle of influence." Of late years, however, evidence has been accumulating that the cone of depression in artesian aquifers at least is to be measured in terms of miles.[5] Meinzer,[6] moreover, has emphasized the large quantities of water that must be derived from storage by many wells and in doing so implied that the extent of the effects of discharge by wells must be very great. The present paper attempts to consider the cone of depression in relation to the quantity of water taken from storage and in the light of its transient and ultimate effects on the aquifer.

[2] Slichter, C. S.: Theoretical investigation of the motion of ground water. U. S. Geol. Surv. 19th Ann. Rept., pt. 2: 359, 1899.

[3] Turneure, F. E., and Russell, H. L.: Public water supplies, 2d ed. John Wiley & Sons, Inc., pp. 279–283, 1908.

[4] Tolman, C. F.: Ground water. McGraw-Hill Book Co., pp. 386–391, 1937.

[5] Thompson, D. G.: Ground-water supplies in the vicinity of Asbury Park. New Jersey Dept. Cons. and Devel. Bull. 35: 24, 25, 1920; Leggette, R. M.: The mutual interference of artesian wells on Long Island, N. Y. Am. Geophys. Union Trans., 1937, pp. 490–494; Barksdale, H. C.: Water supplies from the No. 1 sand in the vicinity of Parlin, N. J. New Jersey State Water Policy Comm. Special Rept. 7: 17, 1937.

[6] Meinzer, O. E.: Compressibility and elasticity of artesian aquifers. ECON. GEOL., 28: 280, 281, 1928.

The term cone of depression seems to have been used in somewhat different senses. As used here, it denotes the geometric solid included between the water table or other piezometric surface after a well has begun discharging and the hypothetical position the water table or other piezometric surface would have had if there had been no discharge by the well. The term as used here is not restricted to the time during which the well is discharging but represents the effects of the discharge at any time after discharge has started, including the time after it has stopped. The term "cone" is a misnomer, as the figure, even in perfectly homogeneous materials, is not a cone, but the term is retained here because of its fixed place in the literature. The vertical distance at any place between the hypothetical uninfluenced position of the piezometric surface and the actual surface after discharge has begun, that is, the lowering due to the discharge, is the drawdown at that place caused by the discharge.

Under this definition, the cone of depression must ultimately include either areas of previous discharge or areas where recharge has previously been rejected or both. All ordinary ground-water bodies, with perhaps a few problematical exceptions, are in a condition of approximate dynamic equilibrium prior to development by wells. All recharge is ultimately discharged by natural means. The implied definite rate of underground flow from points of recharge to points of discharge, according to Darcy's law, necessitates a definite combination of hydraulic gradient and thickness of saturation of the aquifer. Discharge by wells is an additional discharge superimposed on the previously more or less stable hydraulic system. If such discharge is continued indefinitely, it is evident that eventually the aquifer must either receive more water or else the discharge through natural outlets must decrease. Neither of these effects can be accomplished without changing either the gradient of the water table or piezometric surface or the level of the water table in the areas of recharge or discharge, respectively, or, in other words, depressing water levels the entire distance between the well and one or the other of these areas. Even if a well discharges only a short time and

draws its water for the time entirely from storage in the aquifer, eventually the aquifer must either discharge just that much less water through natural outlets or take in that much more water in the intake areas. Eventually, therefore, even in this case, the water table or piezometric surface must be depressed in one or the other of these areas. Hence the cone of depression, as here defined, can have no ultimate limits short of at least one of the hydrologic boundaries of the aquifer. The common practice of estimating the perennial safe yield of an aquifer as equal to the rate of recharge implies the assumption that the natural discharge can be stopped without serious effects at the well field, an assumption that may or may not be justified. These considerations are not important where wells are located close to areas of recharge or natural discharge, as is at least common in humid regions, but they are fundamental in the discussion of effects of wells in most artesian aquifers and in large continuous water-table bodies in the bolsons and plains areas in arid country.

The complete extent of existent cones of depression has not been recognized by observation and perhaps can never be. The drawdown caused by any one well or even any closely spaced group of wells becomes quite small at considerable distances. Some time always elapses between the time when a well begins to discharge and the time when the effects of the discharge are felt at a distance, and this lag confuses the attempt to correlate the lowering of water levels with discharge by distant wells. Finally intermittent recharge, discharge by other nearby wells, or other factors almost always cause fluctuations of water level great enough to mask completely the comparatively small effects of individual distant wells. However, the fact that the cones of distant wells are not individually recognizable does not imply that the sum of the effects of all distant wells is negligible.

Until the cone extends to areas of recharge or natural discharge, it is constantly changing in shape and size. For until recharge and total discharge, natural and by wells, are again equal water must be withdrawn by the wells from storage in the aquifer. In ground-water bodies that have a water table, the extraction from storage results in a lowering of the water table and

consequent drainage of the rock through which the water table falls. Conversely the cone of depression can develop only as fast as the water made available by this drainage can be discharged by the well. A similar effect also occurs in artesian aquifers. In these aquifers extraction from storage occurs because of a compaction and compression of the aquifer following the partial release of the internal hydraulic pressure.[7] The compression of the aquifer is at least largely elastic.[8] Hence discharge by wells from artesian aquifers also entails a measurable period in which the cone of depression is developing. In many areas this is a very significant factor. Meinzer[9] came to the conclusion that artesian wells in the Dakota sandstone after several decades of flow were probably still drawing on storage and hence that water levels were still falling and the cone of depression was still enlarging. Time is an essential dimension of the cone of depression.

These general considerations indicate that in estimating ground-water reserves the effects of wells on water levels elsewhere in the aquifer can be considered merely local only in exceptional cases and that these effects vary in time as well as in space. Under this concept the cone of depression may be considered a pirating agent created by the well to procure water for it, first robbing the aquifer of stored water and finally robbing surface water or areas of transpiration in the localities of recharge or natural discharge. Can this general concept of the cone of depression be made quantitative? Can the effects of one group of wells on other wells in the same aquifer at a particular future time be predicted? Progress toward the solution of these and related problems can be made by considering the effects of discharge by wells in certain idealized types of aquifers capable of mathematical treatment.

THE CONE OF DEPRESSION IN AN IDEALIZED AQUIFER.

In order to treat the problem of well discharge mathematically it is apparently necessary to assume:

[7] Meinzer, O. E.: *Op. cit.*, pp. 263–277.
[8] Idem, pp. 277–280.
[9] Idem, pp. 269–271.

1. The aquifer is homogeneous and hydrologically isotropic.

2. The transmissibility of the aquifer—that is, its ability to transmit water—is constant at all times and places. The transmissibility is expressed quantitatively by a coefficient of transmissibility, defined as the number of gallons of water which will move in one day through a vertical strip of the aquifer 1 foot wide and having the height of the aquifer when the hydraulic gradient is unity. It is the average coefficient of permeability,[10] adjusted for the temperature, multiplied by the thickness of the aquifer in feet.

3. The water taken from storage in any vertical column in the aquifer is always proportional to the lowering of the water table or piezometric surface in the column. This condition is expressed quantitatively by the assumption of a constant coefficient of storage, defined as the volume of water, measured in cubic feet, released from storage in each column of the aquifer having a base 1 foot square and a height equal to the thickness of the aquifer, when the water table or other piezometric surface is lowered 1 foot. In water-table bodies, this coefficient of storage for long periods of pumping is approximately the specific yield [11] of the material through which the water table falls. It is independent of the thickness of the aquifer. In artesian aquifers the coefficient of storage represents the water released from storage by the compression of the aquifer and is presumably directly proportional to the thickness of the aquifer. It is assumed that this coefficient is constant. This assumption implies that under water-table conditions, the water in the material is discharged instantaneously as the water table falls through it. Actually, the drainage is a fairly slow process, as shown by many experiments, including a recent elaborate pumping test by Wenzel.[12] Apparently, however, except for short periods of pumping, the as-

[10] Stearns, N. D.: Laboratory tests on physical properties of water-bearing materials. U. S. Geol. Surv. W. S. P. 596: 148, 149, 1927.

[11] Meinzer, O. E.: Outline of ground-water hydrology. U. S. Geol. Surv. W. S. P. 494: 28, 1923.

[12] Wenzel, L. K.: The Thiem method for determining permeability of water-bearing materials. U. S. Geol. Surv. W. S. P. 679–A: 53–57, 1936.

sumption does not cause serious errors. In artesian aquifers the assumption appears to be at least sensibly realized.

In an ideal aquifer of infinite areal extent conforming to these assumptions, the drawdown at any time and any place would be given by the following equation: [13]

$$v = 114.6F/T \int_z^\infty (e^{-u}/u)du$$

in which

v = drawdown at any point, in feet;
F = rate of discharge of the well, in gallons a minute;
T = coefficient of transmissibility, as defined above;
$z = 1.87r^2s/Tt$;
r = distance from the discharging well to the point where drawdown is to be determined, in feet;
s = coefficient of storage, as defined above;
t = time the well has been discharging, in days.

The definite integral in the equation,

$$\int_z^\infty (e^{-u}/u)du,$$

is the logarithmic integral. It has a definite value for any value of z. Table I gives these values. To compute the drawdown, v, the procedure is (1) compute z, (2) look up in the table the corresponding value of the integral, and (3) multiply this value by 114.6 F/T.

Fig. 1 shows the drawdown at various times at several distances from a discharging well for a particular set of conditions. The assumptions are that the coefficient of transmissibility is 100,000, that the coefficient of storage (specific yield in this aquifer) is 20 per cent or 0.2, and that the well is discharging at the constant rate of 100 gallons a minute. As the drawdown in the ideal aquifer is directly proportional to the rate of discharge, the drawdown for any other rate of discharge, F, would be equal to the drawdown shown on the diagram multiplied by F/100. The values for transmissibility and storage are in a range commonly found in nonartesian aquifers.

[13] Theis, C. V.: The relation between the lowering of the peizometric surface and the rate and duration of discharge of a well using ground-water storage. Am. Geophys. Union Trans., 1935, pp. 519–524.

TABLE I.

VALUES OF THE LOGARITHMIC INTEGRAL, $\int_z^\infty (e^{-u}/u)du$.

(From Table 32, Smithsonian Physical Tables, 8th rev. ed., Smithsonian Inst. Pub. 3171, 1933.)

z	.00	.01	.02	.03	.04	.05	.06	.07	.08	.09
0.0	∞	4.03793	3.35471	2.95912	2.68126	2.46790	2.29531	2.15084	2.02694	1.91874
.1	1.82292	1.73711	1.65954	1.58890	1.52415	1.46446	1.40919	1.35778	1.30980	1.26486
.2	1.22265	1.18290	1.14538	1.10988	1.07624	1.04428	1.01389	.98493	.95731	.93092
.3	.90568	.88151	.85834	.83610	.81475	.79422	.77446	.75544	.73711	.71944
.4	.70238	.68591	.67000	.65461	.63973	.62533	.61139	.59788	.58478	.57209
.5	.55977	.54782	.53622	.52495	.51400	.50336	.49302	.48296	.47317	.46365
.6	.45438	.44535	.43656	.42800	.41965	.41152	.40359	.39585	.38831	.38095
.7	.37377	.36676	.35992	.35324	.34671	.34034	.33412	.32803	.32209	.31628
.8	.31060	.30504	.29961	.29430	.28910	.28402	.27904	.27418	.26941	.26475
.9	.26018	.25571	.25134	.24705	.24285	.23874	.23471	.23076	.22689	.22310

z	.0	.1	.2	.3	.4	.5	.6	.7	.8	.9
1.0	.21938	.18599	.15841	.13545	.11622	.10002	.08631	.07465	.06471	.05620
2.0	.04890	.04261	.03719	.03250	.02844	.02491	.02185	.01918	.01686	.01482
3.0	.01305	.01149	.01013	.00894	.00789	.00697	.00616	.00545	.00482	.00427
4.0	.00378	.00335	.00297	.00263	.00234	.00207	.00184	.00164	.00145	.00129

The value of the integral is given by the series:

$$\int_z^\infty (e^{-u}/u)du = -0.577216 - \log_e z + z - \frac{z^2}{2\cdot 2!} + \frac{z^3}{3\cdot 3!} - \frac{z^4}{4\cdot 4!} + \cdots - (-1)^n \frac{z^n}{n\cdot n!}.$$

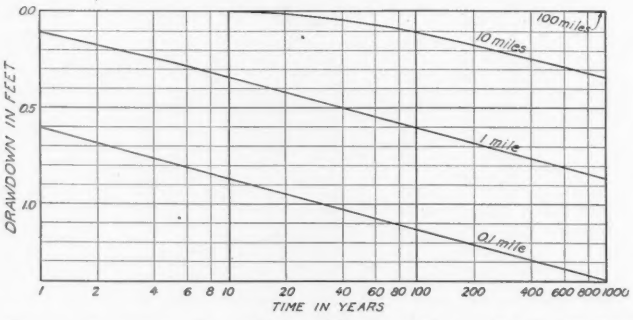

FIG. 1. Drawdown in an ideal aquifer caused by continuous discharge of a well at the rate of 100 gallons a minute. Coefficient of transmissibility is 100,000 and coefficient of storage is 0.2. Distances are measured from discharging well; time from beginning of discharge.

Fig. 2 plots the drawdown against the distance from the discharging well. The successive positions of the cone of depression over most of its extent are nearly parallel. It is because of this parallelism that formulas for the cone of depression based on conditions of equilibrium or steadystate, such as Thiem's

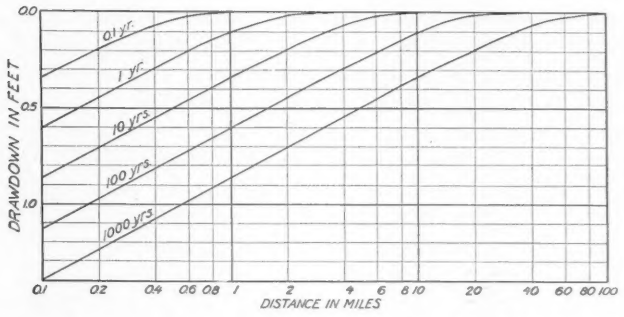

FIG. 2. Drawdown for same conditions as those of Fig. 1 plotted against the distance from the discharging well.

formula,[14] involving as they do only the difference in drawdown at two or more points, can be used even when the cone is far from equilibrium.

Although the numerical values on the figures apply only to the particular assumptions made, the curves themselves are type curves and can be made to apply to any value of coefficients of transmissibility and storage by a change of scale. If the coefficient of storage and the values for time are changed in the same ratio the other values given on the figures remain the same. Because the coefficient of transmissibility occurs twice in the equation the effects of variations in transmissibility are somewhat more difficult to follow. To make the figures applicable to an aquifer of transmissibility, T, it is necessary to divide both the time and drawdown values by T/100,000. To make the figures applicable to a coefficient of storage, s, and a transmissibility, T,

[14] Wenzel, L. K.: *Op. cit.*, pp. 10–23.

divide the values for time by 0.000002 T/s and the values for drawdown by 0.00001 T.

The relation between time and the coefficient of storage demonstrates a significant difference between artesian and nonartesian aquifers. In the ideal aquifer the time necessary to produce a given effect at a given distance from the well is directly proportional to the coefficient of storage. As this coefficient in nonartesian aquifers (specific yield) is of the order of 0.2 and in artesian aquifers is much smaller and seems to be of the order of 0.002 or perhaps 0.0002, the effects of well discharge would travel from 100 to perhaps as much as 1,000 times as fast in the ideal artesian aquifer as in the ideal nonartesian aquifer. These ratios must probably be modified somewhat for actual aquifers, but they serve as a rough quantitative criterion by which to distinguish between effects in the two types of aquifers.

In the ideal aquifer the drawdowns represented by the cone of depression are additive; the same drawdowns occur at equal distances from the well in all directions irrespective of the direction of the normal hydraulic gradient, and the drawdown at any place is the sum of the drawdowns represented by the cones of all wells in the vicinity.

The residual drawdown of the piezometric surface after discharge has stopped can be computed by assuming that the well continued to discharge but that a recharge well of the same capacity was introduced at the same point at the instant discharge stopped.[15] The residual drawdown at any instant is therefore the drawdown due to the discharge from the time discharge started up to the time in question minus the rise due to the recharge well from the time discharge stopped. The latter amount is numerically equal to the drawdown of a discharge well of the same capacity. Thus in Fig. 2 it appears that the drawdown 1 mile from the well after 100 years of discharge would be 0.60 foot, while after 10 years it would be 0.34 foot. The residual drawdown after 90 years of discharge and 10 years of quiescence is the difference between these amounts or 0.26 foot. Beyond a distance of 10 miles the recovery would not yet have begun.

[15] Theis, C. V.: *Op. cit.*, p. 552.

The formula derived here is for the cone of depression in an aquifer of infinite areal extent. As long as the cone does not effectively reach the boundaries of the aquifer—that is, as long as the drawdown at the boundaries of the aquifer is practically zero—the assumption of infinite areal extent is valid for practical application. A discussion of conditions thereafter is beyond the scope of the present paper, but the effects of boundaries can be considered by more elaborate mathematical analyses.[16]

THE CONE OF DEPRESSION IN ACTUAL AQUIFERS.

The mathematical treatment given above applies strictly only to ideal aquifers conforming to the fundamental assumption that they are homogeneous, isotropic, and characterized by constant coefficients of transmissibility and storage. What modifications of the results are needed to make them apply to actual aquifers?

The small local changes in the character of the aquifer probably cause only small and local changes in the character of the cone of depression and are therefore significant only in detailed studies. Of paramount significance are the broad regional changes in transmissibility and storage capacity and changes in these that may take place with continued discharge.

Scattered data seem to indicate that in sandy artesian aquifers the ideal conditions of constant transmissibility and storage capacity are probably closely enough approximated over considerable areas to permit the practical use of the formula for the cone of depression in the ideal aquifer. In a widely extended aquifer equal drawdowns are probably distributed approximately symmetrically about the discharging well. Not enough data are yet available to set accurately the order of magnitude of the coefficient of storage in such aquifers, but preliminary results indicate that it is about 0.000005 for each foot of thickness of the aquifer. This amount is small, and as a consequence the effects of discharge of a well in such an aquifer spread rapidly. However, although the value is small, great quantities of water can

[16] See Muskat, Morris: The flow of homogeneous fluids through porous media. McGraw-Hill Book Co., pp. 621–676, 1937.

be taken from storage in extensive aquifers. Thus, even with this small coefficient of storage, an average lowering of the piezometric surface of 50 feet in an aquifer with an areal extent of 10,000 square miles and a thickness of 100 feet would be accompanied by a withdrawal from storage of 160,000 acre-feet, or 52 billion gallons, of water.

Artesian aquifers of limestone or other soluble rocks may not conform to the ideal aquifer just considered. The processes of solution are likely to have been determined by conditions which resulted in giving a more or less linear pattern to the solution cavities, permitting freer movement in one direction than the other. The aquifer may therefore be anisotropic to the movement of water through it. Discharge of artesian wells in the Roswell Basin, for instance, frequently affects wells east or west of the discharged well more than wells at equal distances north or south of it.

In homogeneous aquifers that have a water table the coefficient of transmissibility decreases in the same ratio as the thickness of saturation decreases. As a first consequence of this the cone is always somewhat deeper than it would be in the ideal aquifer of constant transmissibility. The error increases with the ratio between the drawdown and the initial thickness of saturation of the aquifer. As a second consequence the cone is distorted. In an ideal aquifer of constant transmissibility the only interference with the continued normal movement of the water results from a modification of the hydraulic gradient. In a nonartesian aquifer, however, an additional interference results from the creation of the cone of depression. Water moves into the up-gradient part of the unwatered volume represented by the cone of depression at its normal undisturbed velocity and moves away from the down-gradient part of the unwatered volume at the same velocity. As a consequence, the cone itself drifts down-gradient with the general motion of the ground water.

The process of continuous pumping from such an aquifer may be thought of as occurring in discrete units. The removal of each unit volume of water produces a depression in the water table, at first quite localized, which continually becomes shallower

and broader due to inflow at the edges of the depression. At the same time, this unit cone drifts down-gradient with the general motion of the ground water, becoming of course shallower and broader as it advances. This unit cone is followed by a succession of other unit cones. The drawdown at any point is the sum of the effects of all these cones at that point. The drawdown at a point down-gradient from the well is therefore somewhat greater than the drawdown at a point an equal distance up-gradient from the well. Hence the cone of depression in a non-artesian aquifer is somewhat spoon- or scoop-shaped. Hence also, the natural discharge of such an aquifer will be affected by the pumping sooner than would be the case in an aquifer of constant transmissibility and the areas of recharge will be less quickly affected. However, as the rate of movement of ground water is generally quite slow, these differences may be of small amount.

The application of the theory here outlined to a large continuous water-table body can be shown by a consideration of the probable future effects of pumping in the vicinity of Portales, New Mex., near the western edge of the Staked Plains. Wells in this area draw water from the Tertiary Ogallala formation or from Quaternary sediments hydrologically continuous with the Ogallala. The ground water is recharged by rainfall penetration on the Staked Plains and is largely discharged by seepage and springs at the eastern edge of the Staked Plains, about 120 miles distant. There seems to be no possibility of increasing the recharge by lowering the water table, as the amount available depends on the amount of rainfall that can escape evaporation and transpiration and seep downward. Hence to reach equilibrium an amount of water approximately equivalent to the pumpage must be diverted from the natural discharge at the eastern scarp—that is, the cone of depression must extend that distance. Pumping tests have indicated that the coefficient of transmissibility is about 100,000 in the vicinity of Portales.[17] It is probably no greater in the remainder of the High Plains.[18] The coefficient of storage may

[17] Theis, C. V.: Progress report on the ground water supply of Portales Valley. New Mexico State Eng. 11th Bien. Rept.: 91–95, 1934.

[18] Theis, C. V.: Amount of ground-water recharge in the southern High Plains. Am. Geophys. Union Trans., 1937, p. 567.

be taken as about 20 per cent or 0.20. Fig. 1 indicates that no appreciable effect of the pumping at Portales can be felt at a distance of 120 miles for about 1,000 years. This figure must be modified to consider the rate of drift of the cone of depression in a nonartesian aquifer. However, with a coefficient of permeability of 1,000 and a hydraulic gradient of 0.002, or about 10 feet to the mile, the normal velocity of the ground water, and hence the cone of depression, is only about 500 feet a year, if a specific yield of 20 per cent is assumed. Hence the time taken for the cone to drift the 120 miles to the scarp would be about 1,200 years. It appears probable, therefore, that the wells at Portales and other wells far from the localities of natural discharge of such large nonartesian water bodies must pump from storage in the aquifer for many generations. However, although most of the water taken may be withdrawn from storage the withdrawal will be spread over a very large area, so that the average drawdown will be small and the effects at the wells themselves may not be great if the wells are properly spaced.

Replenishment of any aquifer as a whole can begin only after the cone of depression effectively reaches the areas of natural discharge or of rejected recharge. The time when this occurs is independent of both the rate of discharge and the continuity of discharge of the wells. Apparently in extensive water-table aquifers, such as the Ogallala of the southern High Plains, this time is to be measured in centuries. Although in a geologic sense such a ground-water reserve is replenishable, it does not appear to be so in the human sense.

U. S. GEOLOGICAL SURVEY,
WASHINGTON, D. C.,
Aug. 9, 1938.

DISCUSSION AND COMMUNICATIONS

HISTORY OF THE THEORY OF SECONDARY SULPHIDE ENRICHMENT

Sir: The historical contribution by J. S. Brown (this journal, 33: 211–215, 1938) on the early recognition of the zone of secondary sulphide enrichment is highly interesting. Nevertheless, the citations do not give the earliest data for such recognition. In his excellent publication, W. H. Emmons[1] mentioned the similar views of J. D. Whitney expressed in 1854–55. Whitney described the narrow beds of rich black copper sulphide occurring between the oxidized ores and the leaner chalcopyrite-magnetite zone at Ducktown, Tenn., and designated them as "gossan."

The earliest one, however, to my knowledge, who completely and definitely recognized the zone of rich secondary copper ores was the German mining man and mineralogist, Martin Websky.[2]

Websky published an article[3] entitled "The geognostic relations of the ore deposits of Kupferberg and Rudelstadt in Silesia." In the locality cited, there were at the water table large masses of chalcocite and covellite, which Websky definitely regarded as secondary alteration products of chalcopyrite in the weathering zone. The observations and conclusions of Websky were mentioned and discussed in detail by Gustav Bischof (1792–1870, one time professor at Bonn), in assembling the great "Lehrbuch der chemischen und physikalischen Geologie."[4] Bischof agreed entirely with the conclusions of Websky.

[1] U. S. Geol. Surv. Bull. 625: 12–13, 1917.

[2] Christian Friedrich Martin Websky, born in 1824 in Silesia, was first a mining engineer, mine valuation engineer, and mine advisor in Breslau; then became Instructor at the University of Breslau; and was later appointed Professor of Mineralogy first at Breslau and then at Berlin. He died in Berlin in 1886. Websky was an exceptionally careful observer and one of the ablest mineralogists of all time.

[3] Die Geognostischen Verhaeltnisse der Erzlagerstaetten von Kupferberg und Rudelstadt in Schlesien. Zeit. der Deutsch. Geol. Gesell. 5: 373–438, 1852.

[4] See vol. II, 3 Abt., pp. 1922 et seq, 1855.

Another early observer of sulphides due to secondary enrichment should be mentioned. A. Knop (1828–1893, once professor at Karlsruhe) published in 1861 a work entitled "On Copper Ore Deposits in Lesser Namaqualand and Damara, Southwestafrica—a Contribution to the Genesis of the Copper Ores."[5] Here the author describes in great detail the secondary alteration of chalcopyrite into bornite, chalcocite, and covellite in the weathered zone. His concluding sentence, quoted below, shows that, even at this early date, he contended for a more widespread application of his observations:

> It is highly probable that relations similar to those governing the copper ores of Africa can be observed in many other parts of the earth, if once the attention of mining men is specially directed to the genesis of these ores.

The further development and the ensuing studies are of course well known. About 1890 Gonzalo ÿ Tarin recognized the secondary copper sulphides in the Huelva district. The first general account of weathering of ore deposits is to be credited to R. A. F. Penrose.[6] Thereafter came the work of L. de Launay in 1897 and finally in 1901 the basic studies of S. F. Emmons, W. H. Weed, and C. R. Van Hise.

I have already presented the historical development of the theory in my detailed work on "Oxidation and Enrichment Zones of the Sulphide Ore Deposits" (Die Oxydations- und Zementations-zone der sulfidischen Erzlagerstaetten") in Fortschritte der Mineral., vol. 9, pp. 67–160, 1924.

HANS SCHNEIDERHÖHN.

UNIVERSITY OF FREIBURG, GERMANY,
Aug. 23, 1938.

[5] Neues Jahrb. f. Mineral, pp. 513–550, 1861.

[6] The superficial alteration of ore deposits. Jour. Geol., 2: 288–307, 1894.

REVIEWS

Steinsalz und Kalisalze. Geologie. By FRANZ LOTZE. Vol. 3, Pt. 1 of "Die Wichtigsten Lagerstätten der 'Nicht-Erze,'" by O. Stutzer, pp. 936; figs. 353. Gebrüder Borntraeger, Berlin. 1938. Price RM 84.

Steinsalz und Kalisalze. By ERNST FULDA. Vol. 3, Pt. 2 of "Die Lagerstätten der nutzbaren Mineralien und Gesteine," by Beyschlag, Krusch and Vogt, pp. 240; figs. 94. Ferdinand Enke, Stuttgart. Price RM 18.20.

Zechstein. By ERNST FULDA, Editor. Part of "Handbuch der vergleichenden Stratigraphie Deutschlands," Preuss. geol. Land., pp. 409; figs. 100. Gebrüder Borntraeger, Berlin, 1935.

Although the last of these three books appeared in 1935, so large a part of it is closely related to the other two that it seems appropriate to review the three at one time. Together they form a full, if not a complete account of the geology of salt and of the salt deposits of the world. The last named deals only with those of Germany and in the other two a large proportion of the space naturally is given to the German deposits, but as they are the best known of the world, this is an advantage.

Geologists are fortunate in having had the authorship of the first-named volume fall into the hands of Lotze. For some time he was connected with the University of Göttingen, almost in the heart of the German salt and potash region, yet he has also had a wide and varied experience in general structural geology. He thus brings to the discussion of salt an appreciation of its broader geological bearings and a love for the study of it, charmingly expressed in his preface where he refers to salt as "the livest and most temperamental of rocks."

It is hard to conceive how a man active in so many directions could in a few years produce so masterly and thorough a work. Throughout, one senses that a vast literature has not only been gone over, but assimilated, to be drawn on wherever it can contribute anything relevant.

However, the book is not a mere compilation of the work of others. Every phase of the geology of salt is analyzed and interpreted on the basis of the literature and of the author's own experience, and even though one may not be ready to accept a certain conclusion, the problem and the different possible interpretations are so clearly and logically presented and so thoroughly documented that the reader is well prepared to form his own opinion.

Part I (pp. 1–321) treats of the general geology of salt, with which gypsum and anhydrite are included. Part II (pp. 322–824) relates to regional salt geology. Of this second part, more than one-third (pp. 379–561), are devoted essentially to the famous Permian salt deposits of Germany.

In the first part all phases of the geology of salt are treated. There is a full discussion of the origin of salt deposits on the basis of modern examples grouped in different types and this is followed by an enumeration of occurrences of salt according to geologic periods and by an attempt to interpret the origin of fossil deposits on the basis of an analysis of their characters. The last named is perhaps the least convincing part of the book. Not that the presentation is not clear and critical, but our present knowledge is not sufficient. Compared with other sedimentary rocks, the composition of salt deposits is so complex, the factors that may enter into their origin are so numerous, and they are so mobile both chemically and physically, that progress in reconstructing the conditions of their origin must necessarily be slow. The complexity of geologic conditions affecting salt deposition is well illustrated, for instance, by the Dead Sea and Salton Sink lake as described in this book.

In connection with the chronologic enumeration of salt deposits, Lotze presents and discusses on pages 154 to 159, a series of maps showing the zonal distribution of salt deposits in the northern hemisphere in successive geologic periods. These maps reveal a fairly constant migration of the salt zone from the pole towards the equator. As present day salt formation in the northern hemisphere takes place mainly in the arid belt north of the equator, migration of the poles is indicated. But Lotze mentions other possible interpretations and leaves the problem open.

Of general interest to geologists is also the discussion on pages 162–167 of a cyclic or rhythmic succession in salt beds.

The discussion of the origin of salt deposits is followed by a discussion of their metamorphism and of the phenomena, so significant for geology in general, of their tectonics, of which salt stocks (diapirs) are probably the best known but not the only important manifestation. However, the wide distribution of salt stocks revealed by the second part of the book, which will probably be surprising to most geologists, serves well to emphasize the importance of salt tectonics in general.

The first part closes with a section dealing with the destruction of salt deposits, of which underground solution (called "Subrosion" by Germans) is the most complex geologically.

In the second part, dealing with salt deposits regionally, the factors presented in the first part of the book are discussed in relation to the different regions, and thus much additional light is thrown on these factors. The individuality of most deposits, even of those closely related

geologically and geographically, is one of the striking facts brought out. The section also includes brief technical data on individual mines, drilling, reserves, and so on. In this connection special attention should be drawn to an enumeration of potash occurrences of the world on page 148 of the first part, and to the statement on page 127 that all large salt deposits contain some potash.

Especially rich in broadly illuminating facts and viewpoints is the discussion of the stratigraphy of the salt of the classical Stassfurt region, where the brilliant work of Tinnes in differentiating guide horizons of common salt of great lateral extent, that pass unchanged through different zones of potash salts, has made it possible to establish a true stratigraphy and to recognize the effects of metamorphism on the salts (Fig. 249, p. 461).

Regarding the relative part played by orogenic forces on the one hand, and epeirogenic forces or the weight of overlying material on the other, in producing salt stocks or other salt intrusions, Lotze ultimately comes to full recognition of the part played by gravity, but not without casting a lingering look behind (pp. 275–283). Thus, the first paragraph of the section in question, and the last two paragraphs on page 283, seem weighted on diametrically opposite sides of the question. It is difficult also to picture the process (p. 276) by which orogenic forces could act on salt before they had affected any of the enclosing beds. Likewise, it is not obvious why, if it is assumed that gravitative rise of the salt stock of Salinas de Añana caused thinning of Tertiary strata above it, orogenic forces should be assumed to have caused the stock to push up those same beds.

In this and other instances, Lotze seems reluctant to abandon orogenesis as the cause of any given movement of salt, yet in his formal conclusions on page 283 he does do full justice to gravitative or hydrostatic rise.

Restrained as are Lotze's statements about recognizable tectonic lines in the Gulf Coast salt dome region of the United States (p. 277), it is questionable whether he does not still go too far. In only a few places, he says, can clearer lines be recognized, elsewhere they can be reconstructed "with difficulty." Experience seems to indicate that it would be better to say, "only with abundant imagination."

Though Lotze's treatment of the United States is remarkably full and accurate, there are a few statements of doubtful validity. Thus, he misinterprets the authors he cites in assuming that it may be taken as *fairly certain* that the salt of the Gulf Coast salt stocks is the same as what he calls the Red Bed salt of Arkansas (p. 738), and he is probably particularly misled in assuming that the so-called sandstone dyke in the salt of Avery Island supports that conclusion.

At Searles Lake, borax is a by-product of potash, rather than, as Lotze states (p. 746), the reverse.

However, these might be called errors of emphasis rather than actual misstatements, and as the presentation is clear and the literature is amply cited, they do not detract appreciably from the value of the book.

The book is richly illustrated with both half tones and line cuts, most of them very pertinent, and many of them apparently original with the author and prepared especially for this book. It is unfortunate, however, that so often authors who take the greatest pains to make their text clear and accurate begrudge the time necessary to make their illustrations complete and readily grasped. Probably Lotze cannot be held responsible for the inadequate lettering and description of such half-tones as Fig. 168, p. 255; 170, p. 257; and 172, p. 258, as they are reproduced from the work of another. But there seems little justification for the lack of coördination between the closely related Figs. 235 to 237, and for their poor correlation with the text. Not enough towns and other geographic features are shown. (To get some idea of general geographic relations, one must refer to Fig. 231, p. 401, or to an atlas.) Although they are all on essentially the same base, they do not show the same towns; the stippled areas are not properly explained and named on any of the maps. Fuhrbach (p. 421), is not shown even in Stieler's atlas. Probably this area is so familiar to a German that he does not need the missing details, but a foreigner who wants to read the related text intelligently, is compelled to supplement the map by independent geologic and geographic research.

Readers who are not in the habit of reading lists of errata are warned to do so before studying Fig. 86, p. 180, the diagram illustrating the bar theory.

As Lotze himself points out, the treatment of different parts of the book is not equal. Thus, towards the end, the book reached such dimensions that Africa and Asia have not received the consideration they deserve.

All these are minor defects, however, and it can only be reiterated that every geologist will find Lotze's book rich in information and in illuminating ideas. Special gratitude is also due the author for his splendid index covering 112 pages.

Fulda, the author of the second and author and editor of the third book, is salt expert of the Prussian geological survey, and consequently brings to his work detailed knowledge of German salt deposits. Though his book on salt covers the same general fields as Lotze's—being like Lotze's divided into a general part (some 60 pages) in which all details of salt geology from origin through tectonics, metamorphism, solution, and so on, are treated, and a regional part (some 175 pages, of which some 50 go to Germany) yet the emphasis is naturally different.

The most striking difference is in the fuller treatment of the mineralogy and of the mineralogic stratigraphy. In the series in which Lotze's book is published, the mineralogy of salt is to be treated in a separate volume.

Fulda tends also to attribute more of the characters of salt deposits to the conditions of their origin (the older idea) and less to metamorphism, than do some other authors. Fulda's work also differs from Lotze's in giving some history and statistics of production. A special feature is the discussion of radioactivity of salt and its relation to the origin of blue rock salt. Though Lotze acknowledges the help of Polutoff in writing up the Russian deposits, Polutoff is given as actually the author of the section dealing with the Russian and Siberian deposits in Fulda's book, which may be regarded as an advantage; an advantage, however, which, as indicated by relative space, falls mainly to the Siberian deposits.

Fulda's book is much shorter than Lotze's, and that may justify to a certain extent a more fragmentary, less rounded treatment; but one feels, throughout, a less broad approach and a less balanced judgment. Both Lotze and Fulda accept as fundamental in the origin of great salt deposits, the concept of intermittent supply of sea water to a vast marginal basin, and both mention the Grossflutentheorie (giant tide theory) of Wilfarth[1, 2] which is based on George Darwin's theory that the moon was formerly much nearer the earth and that tides must then have had much greater range.[3] This "Grossflutentheorie" may have much in its favor and may be an important contribution to the solution of the problem of the origin of large salt deposits, but to say, as Fulda does on pages 119 and 124, that from the point of view of the "Grossflutentheorie," the "Haselgebirge" *must* be regarded as a primary deposit, seems rather premature. The Haselgebirge is a peculiar breccia of anhydrite and clay in salt. Generally it has been regarded as secondary and recent students, including Schauberger, whom Fulda does not cite in that connection on page 124, have assumed a tectonic origin.

Few geologists, probably, will agree with Fulda's assumption (p. 227) that the oil around American Gulf Coast salt stocks probably originated below the salt.

[1] Wilfarth, M., Sedimentationsprobleme in der Germanischen Senke zur Perm- und Triaszeit, Geologische Rundschau, 24: 349–377, 1933; Strömungserscheinungen im Wellenkalkmeer, Ztschr. Deut. Geol. Gesel, 86: 265–285, 1934.

[2] To the references, in Lotze's index, to the work of Wilfarth should be added pp. 163–164, where Wilfarth's theory is first explained.

[3] As Fulda notes (p. 221), A. C. Lane, in 1895, attributed certain features of the salt deposits of Michigan to great tides, though he did not attempt to account for these tides. Fulda's reference to Lane should be to part 2 of the volume he refers to.

Fulda's book on the Zechstein is especially valuable for its detailed description, with many cross sections, of the individual occurrences of salt, of which a large proportion are, of course, salt stocks. Thus the tectonics of German salt deposits are more fully treated here than in either of the other two books, so that Lotze himself refers his readers to Fulda's book for this phase of the subject. The stratigraphy of the formations involved, both the saline and non-saline, is also treated more in detail than in the other two books. About 55 pages deal with the origin of salt deposits, but Fulda had at that time not yet adopted the "Grossfluten-theorie." Some 180 pages deal mainly with individual salt occurrences.

Much of the book, of course, deals with non-saline formations, but among these there are some that are of more special interest to economic geologists. Foremost among them is the Kupferschiefer. Petroleum also receives attention throughout the book. Among other economic products, iron ore and barite are listed on page 96. The book contains, in a pocket, a very clear and useful geologic map showing salt domes and oil fields.

Regarding the origin of the gypsum-anhydrite cap rock, Fulda seems quite confused, a result, probably, of the fact that on German salt stocks it is known mostly as gypsum. On page 42 of his salt book, and page 51 of that on the Zechstein, he recognizes the participation, in its origin, of disseminated particles of anhydrite freed by solution of the salt, but introduces the strange idea that katatectic banding is the result of gypsification of this residual anhydrite instead of a primary character. On page 227 of his salt book, he rejects the concept of origin by residual accumulation and goes back to the assumption that the anhydrite cap of Gulf Coast salt stocks is a bed of anhydrite pushed up on top of the salt. One would also like to know the author's reasons for concluding that most of the solution that produced the salt table of salt stocks probably took place subaerially. But the greatest confusion regarding not only the origin of the gypsum anhydrite cap, but of the relations of the salt table as well, is indicated by many of his cross-sections of salt stocks, especially in the volume on the Zechstein. Unfortunately, these cross-sections do not apparently show all the drill holes and mine workings on which they are based, so that it is not possible to be sure whether any given detail is purely hypothetical or based on observation. In general, however, Fulda apparently assumed that the gypsum-anhydrite cap of German salt stocks is formed only from thick beds of anhydrite, especially from the "Hauptanhydrit." In fact, on page 117 of the Zechstein volume, he definitely says that it is presumably formed mainly in that way, though on page 51, as pointed out above, he recognizes the participation of finely disseminated anhydrite.

A few figures chosen at random from the Zechstein volume to illustrate this confusion concerning the gypsum-anhydrite cap rock are 28, p. 150; 29, p. 152; 58, p. 219; and 66, p. 270. Even if one is willing to believe in the complete absence of a gypsum-anhydrite cap on the salt table of the salt beds flanking the "Hauptanhydrit" in these cross-sections, such relations as those expecially conspicuously illustrated in figure 58, where the Buntsandstein overlying the salt is also shown in parallel extension of it above the salt table, are nothing less than ridiculous. If the salt table was formed subaerially, it is in fact difficult to explain the exclusive presence above the salt table, even in a collapsed condition, of the bed stratigraphically next above the salt sequence.

On the other hand, contrasting with figures showing this improbable restriction of the gypsum-anhydrite cap to direct association with the "Hauptanhydrit" are figures like 38, p. 170; 42, p. 178; 44, p. 188; and 50, p. 195, in which the gypsum-anhydrite cap occurs in no association, or at least no direct association with a massive bed of anydrite.

Since gypsum forms from anhydrite near the surface, and in turn goes over to anhydrite when deeply buried, it has always been a problem to know in which form sedimentary calcium sulphate was deposited. It is, therefore, worth noting that Lotze (p. 183) believes that most of it was deposited as gypsum, whereas Fulda (Zechstein, p. 26) states that it formed as anhydrite.

Whichever conclusion one may be inclined to accept, the belief that anhydrite was the original form cannot be substantiated by the observation (Fulda, Zechstein, p. 57) that enterolithic structure, commonly supposed to be due to gypsification, is found in anhydrite; for just as anhydrite goes over to gypsum near the surface of the earth, so gypsum can go over to anhydrite on being deeply buried.

The ready solubility of salt makes it an especially sensitive recorder of the effects of ground water and the discussion of this phase of the geology of salt in all three books should therefore be of general interest to geologists.

The attention of those interested in varves should be drawn to the discussion by both authors of the so-called annual layers in salt deposits, marked by dark concentrations of disseminated anhydrite. Fulda (Zechstein, p. 136) concludes that they are not annual but Lotze (pp. 163–167) in a more extensive discussion of the problem, questions this conclusion.

All three books have full bibliographies segregated by topics.

For those who must choose, Lotze's book must be recommended above all as the broadest and most complete. Fulda's ideas on most of the topics involved can be learned by following up the references under his name in Lotze's index, and they will be found in Lotze's book in a critical setting and with other ideas on the same topic.

For details on German Permian salt deposits, and especially on salt stocks, Fulda's book on the Zechstein is of special value.

MARCUS I. GOLDMAN.

U. S. GEOLOGICAL SURVEY,
WASHINGTON, D. C.,
October 13, 1938.

Outlines of Geomorphology. By S. W. WOOLDRIDGE AND R. S. MORGAN. Pp. xxi + 445, Figs. 272, Pl. 1. Longmans, Green & Co., London and New York, 1938. Price $4.50.

This volume, which was published in England under the title of "The Physical Basis of Geography," is a discussion of the more important aspects of geomorphology. It was designed as "a guide to a difficult and scattered literature with which students of both geography and geology are concerned." It deals with certain aspects of geophysics and structural geology and with landforms due to erosion. The latter portion of the book is largely concerned with the processes and results formerly denominated "Physiography." It is based mainly on the conception of the cycle of erosion initiated by Prof. W. M. Davis. The first part summarizes and discusses the theories proposed to account for the geological processes that gave rise to surfaces that were subsequently subjected to the erosional processes to produce successive landscapes. In dealing with this part of their subject the authors "sought to beat a tentative path through a veritable jungle of conflicting hypotheses, adopting a point of view favorable to 'continental drift,' sub-crustal thermal cycles, and certain modern theories of mountain-building." Their treatment of "conflicting hypotheses" is fair and clear, and is not confusing to the reader because the authors accept certain of the hypotheses as more probable than others and give the reasons for their decisions. The discussion of continental drift and of mountains and their origin are excellent examples of this treatment.

The book is modern in every respect. It is readable, is well illustrated and is not so elementary as to be uninteresting to college students. It is provided with an excellent selected bibliography and a rather comprehensive index. The illustrations are good, though it might have been more satisfactory to the American reader if a larger proportion of them had been taken from American sources. However the volume is well worth the consideration of students everywhere.

W. S. BAYLEY.

The Principles of Soil Science. By A. A. J. DE SIGMUND. Translated by A. B. Yolland. Pp. xiv + 362, Figs. 34, Pl. 4. Thos. Murby & Co., London. 1938. Cloth, 9½ × 6¼. Price 22s. 6d.

"The Principles of Soil Science" is the translation of a condensation of the author's larger volume "Soil Science," which has been shortened by omitting the sections on soil physics and microbiology, which he feels are satisfactorily treated in English books. In the "Foreword" Russell of the Rothamsted Experiment Station writes that "the main theme of the book is the presentation and discussion of the system of soil classification already known internationally by the author's name, but never before described in detail in an English publication," and which, consequently "has not received from English and American investigators the careful scrutiny which it merits as a positive contribution towards the advancement of soil science."

Although there may be some criticism of the details of de Sigmund's system of classification, which is founded essentially on the chemical composition of the soil, it furnishes "a universal classification based on strictly scientific principles" for discussion by the experts in soil science of all countries.

The volume is divided into four parts—genetics, agronomy, soil systematics and principles of soil cartography. Of these soil systematics occupies by far the greatest portion of the book. It describes twenty-five soil orders including forty-seven main types. It is impracticable to review these in detail in the present place. It is enough to say that the descriptions and discussions are clear and easily understandable and that the translation is in good idiomatic English.

W. S. BAYLEY.

The Silver Magnet. By GRANT SHEPHERD. Pp. 302. Illus. 14. E. P. Dutton & Co., New York. 1938. Price $3.00.

This book contains a matter-of-fact account of the author's experiences in the silver mining camp at Batopilas in the Sierra Madre Mountains near Chihuahua, Mexico. It begins with the arrival of the author as a boy of 5 years at the old mining camp with his father and family, after a trip by rail, wagon and pack train from Washington, D. C., to a point 300 miles southwest of Chihuahua. For many years the family was isolated by distance from its friends and relatives in the United States and the youngsters were brought up very much as were their Mexican companions.

The book recounts the author's experiences, first as a young boy, then as a youth working in and about the mines and the finally as the superintendent of one of the important ones. It is not a continuous record but

rather is a series of descriptions of various episodes in his life which throw light on the conditions under which he lived in a wild environment in the midst of a foreign community. There are related many incidents that should be of great interest to one who has never worked under conditions where hardships are so common that they are not regarded as hardships but rather as the usual concomitants of life.

It is interesting to note at how early an age the author and his brothers accepted administrative responsibilities in the mines and how well they managed their labor, while at the same time living with it on democratic terms.

The volume is well worth reading by those who enjoy the descriptions of life on extreme frontiers.

BOOKS RECEIVED.

J. D. BATEMAN.

Grafita No Estado de Minas Gerais. O. H. LEONARDOS. Pp. 24; pls. 2. Serv. de Fomento da Prod. Min., Avul. 26, Rio de Janeiro, 1938.

Leis Especiais do Petroleo, Brazil. Pp. 15. Serv. de Fomento da Prod. Min., Avul. 27, Rio de Janeiro, 1938. *New legislation affecting the petroleum industry.*

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Mines of the Southern Mother Lode Region, Part 1—Calaveras County, California. C. E. JULIHN AND F. W. HORTON. Pp. 140; figs. 34. U. S. Bur. Mines Bull. 413, 1938. Price, 30 cts. *One of a series of mineral industry surveys of the U. S.*

Composition of Coal Tar and Light Oil. C. H. FISHER. Pp. 70. U. S. Bur. Mines Bull. 412, 1938. Price, 15 cts.

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The Petroleum Engineer. Vol. X, No. 1, Oct., 1938. Pp. 250. Dallas, Texas. $2, annually. *This 10th volume contains a special foreign section with 18 papers relating to occurrence, production and drilling in India, Burma, East Indies, Borneo, Bahrein Saudi Arabia, Kuwait, Iraq, Iran, Java, Sumatra, Assam, Punjab, Russia, Roumania and Netherlands Indies.*

SOCIETY OF ECONOMIC GEOLOGISTS

The Society of Economic Geologists will hold its annual meeting with the Mining Geology section of the A. I. M. M. E. on Feb. 13–16 at the annual meeting of the Institute in New York City. One session will be devoted to a discussion of the structural control of ore bodies, continuing similar discussions previously held. Papers on this or on any subject relating to economic geology will be welcomed.

It is planned to publish abstracts of papers to be given in the January–February issue of ECONOMIC GEOLOGY and reprints of the abstracts will be available at the meetings.

Titles and abstracts from members may be sent to Philip Krieger, Department of Geology, Columbia University, New York, N. Y. The closing date for such abstracts to be printed in the Bulletin is January 10. Titles from members of the Institute may be sent to Charles H. Behre, Jr., Department of Geology, Northwestern University, Evanston, Illinois.

The annual dinner of the Society of Economic Geologists will be held at the Harvard Club, 27 West 44th Street, New York City, at 7:00 P.M. on Tuesday, February 14th, at which time the Presidential address will be given.

The Society will hold an informal meeting in conjunction with the Geological Society of America, which celebrates its 50th Anniversary in New York, December 28–30.

SCIENTIFIC NOTES AND NEWS

A GEOLOGICAL INSTITUTE has been founded in CARACAS, VENEZUELA and on September 15 began classes with 24 students. Training will be in general, petroleum and mining geology. Courses will be offered in general geology, mineralogy, economic geology, petrology, paleontology, stratigraphy, sedimentation and field geology and basic courses in sciences and languages. A building, necessary equipment, research laboratories and preparation rooms have been provided. The course is four years, leading to the B.S. degree. Six scholarships are available to students of North and South American countries.

THE NEW ENGLAND INTERCOLLEGIATE GEOLOGICAL ASSOCIATION held its 34th Fall Excursion in the central Vermont marble belt October 14–16 under the direction of George W. Bain. An excellent Guidebook was prepared depicting many stratigraphic, structural and economic features.

LESTER S. THOMPSON, until recently conducting geological exploration for Amiranian Oil Co. in Iran, is now connected with the Iraq Petroleum Co., City Gate House, Finsbury Square, London.

VICTOR M. LOPEZ of the Servicio Tecnico de Geologia y Mineria del Ministerio de Fomenta in Caracas, Venezuela, was recently granted the degree of mining engineer by the Universidad Central de Venezuela.

OLIN G. BELL has been engaged as Gulf Coast production geologist for Humble Oil and Refining Co.

R. L. LOOFBOUROW is now with the Reliance Mining Co. at Manhattan, Nevada.

GLEN FRANCIS BROWN, who has been with the Bureau of Mines in Manila, is now stationed in University, Miss., with the U. S. Geological Survey.

JULIUS KRUTTSCHNITT, managing director of Mount Isa Mines, Ltd., is the new president of the Australian Institute of Mining and Metallurgy.

MARK C. MALAMPHY, geophysicist, who joined the organization of Hans Lundberg Ltd. after his return from South America last year, will specialize on geophysical problems of the Appalachian Region.

JOHN D. GALLOWAY, who has been associated with Reward Mining Co. is now representative in British Columbia for the Mining Corporation of Canada.

GEORGE A. BROOMELL, geologist for the Soriano Interests, has returned from New York to Musoma, Tanganyika, British E. Africa.

PRESTON LOCKE was accidentally drowned from his yacht at Vancouver, B. C., on October 5th.

CHARLES E. SCOTT, formerly general superintendent of the American Metal Company of Texas, has been made assistant general manager of the Compañia Minera de Peñoles of S. America. His address is now Apartado 251, Monterrey, N. L., Mexico.

ERNEST R. LILLEY, Professor of Economic Geology at New York University, has been appointed chairman of the Committee of the Annotated Bibliography of Economic Geology to succeed Professor Waldemar Lindgren who initiated the Bibliography and served as its chairman for the first 10 volumes just ended.

E. S. MOORE of the University of Toronto has returned from a trip to Africa where he spent the past summer visiting mines in South Africa and Northern and Southern Rhodesia.

HOWLAND BANCROFT, Brazilian consultant, has returned to New York from Rio de Janeiro.

FORBES WILSON has been recently appointed general superintendent for the Timmins Ochali Mining Co. of Yarumal, Antioquia, Colombia.

FREDERICK G. CLAPP delivered a lecture entitled: "Recent Explorations in Eastern Iran" before a joint session of the Iran Society and the Royal Central Asian Society in Royal Societies Hall, Burlington House, London, on October 28th.

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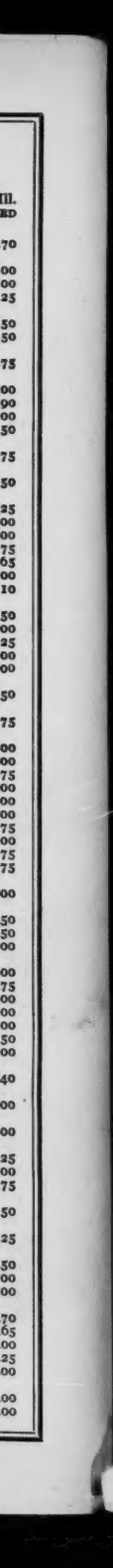
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